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COMPOSITION OF THE MELILITE GROUP

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INTRODUCTION

Many workers have made studies of the interesting series of minerals comprising the melilite group in the attempt to get satisfactory end members to explain their composition. Schaller¹ in 1916, summarized the previous work and contributed a careful study of all the existing analyses. Later Ferguson and Buddington² prepared a series of synthetic equivalents of the members of the melilite group in which they showed that $\text{Ca}_2\text{MgSi}_2\text{O}_7$ and $\text{Ca}_2\text{Al}_2\text{SiO}_7$ were completely miscible. Then Buddington³ using these two end members and adding the sarcolite molecules, as interpreted by Schaller, made an elaborate study of synthetic mixtures, with only partial verification of the validity of the sarcolite molecules, as given by Schaller. In 1924 Winchell,⁴ for the first time considered the melilite group from the standpoint of volume isomorphism. He recognized the fact that all melilites are essentially of the formula R_5O_7 with R equal to all the bases, including silicon. He also pointed out the improbability that such dissimilar molecules as Schaller and Buddington had suggested could enter into the melilite composition. In his study in addition to the åkermanite and gehlenite members, as suggested by Buddington he proposed the molecules $\text{Na}_2\text{Si}_3\text{O}_7$ and $\text{Ca}_3\text{Si}_2\text{O}_7$, in agreement with his general R_5O_7 formula.

The study of this group was undertaken by the author in the belief that the theory proposed by Winchell was essentially correct but that the simplification of the composition of the group did not adequately explain the analyses. A study by the author on all the available analyses has led to a somewhat different interpretation of the group, due mainly to the fact that more information on the composition of the silicates has recently been disclosed by

x-ray structure studies on some of the more complex minerals. W. Wahl⁵ and B. Gossner⁶ have in recent papers attempted to write structural formulae for this series. It is the opinion of the author that a more direct means of studying the structure of the silicates is available in the x-ray methods of Bragg⁷ and his school, who are at this time making a careful study of such complex silicates as the amphiboles and pyroxenes, topaz, andalusite, sillimanite, etc., with results that seem to justify the treatment accorded the melilites in this paper.

All of the available analyses of melilites have been collected in Table I. With each analysis is given the calculated composition according to the theory proposed in this paper, and also the differences between the reduced analyses and the theoretical composition. There is also given, for comparison, the discrepancies obtained by the use of Schaller's theoretical end members.

In Table II the analyses are given in terms of the atomic composition with a constant number of oxygen atoms. The numbers refer to the analyses numbers in Table I. Both the found and theoretical atomic compositions are given in the same table to facilitate comparison. A detailed discussion of this table is given in the theoretical part of the paper.

Table III gives the molecular percentage composition of the analyses according to the end members adopted in this paper.

I. THEORY

All the available analyses (Table I) of members of the melilite group have been reduced to their atomic ratios on the basis of 70 oxygen atoms, as has been suggested by Winchell's R_5O_7 formula, and by the two most likely end members, åkermanite $Ca_2MgSi_2O_7$ and gehlenite $Ca_2Al_2SiO_7$. That this is a fundamentally sound procedure, is verified, in the author's opinion, by the results obtained in Table II which is a list of the analyses in this manner. If the artificial åkermanite, gehlenite series is a series corresponding to the natural melilites, then there is a constant value for $Mg+Al+Si$ of 30 atoms to 70 atoms of oxygen, and any intermediate members having Mg , Al , Si , will have as their total 30 atoms. This fact is amply borne out by Table II. The sum of $Mg+Al+Si$ is in all cases very close to 30 when the oxygen atoms are taken as 70. The deviation from 30 is significant in but a very few analyses and is within the limits of error in most.

TABLE I. ANALYSES OF MELILITES.

	CaO	Na ₂ O (K ₂ O)	MgO	Al ₂ O ₃	SiO ₂	Fe ₂ O ₃	FeO etc.	Total	REFERENCES
1. Åkermanite, anal. (a)	39.62		13.38	1.09	46.70		0.12	100.79	F. Zambonini, <i>Miner. Vesuv.</i> , p. 255, 1910. From Vesuvius.
reduced av. (b)	39.30		13.30	.96	46.55			100.23	
calc.	39.27		13.31	1.02	46.40				
diff.	39.70		13.29	.77	46.24				
diff. by W. T. S.	+0.43		-0.02	-0.25	-0.16				
	-0.42		-0.00	+0.08	+0.34				
2. Åkermanite, anal. (Humboldtite A.)	35.58	2.95	7.60	11.37	39.86	0.50	1.78 H ₂ O = .59	100.26	H. S. Washington, Analyst. <i>Am. J. Sc.</i> , 5th Ser., Vol. 3, 1922, p. 77. Monte Somma.
reduced	35.69	2.98	9.44	11.91	39.98				
calc.	36.44	2.76	8.68	11.38	40.74				
diff.	+0.75	-0.22	-0.76	-0.53	-0.76				
diff. by Buddington	+1.13		-0.35	-0.69	+0.02				
3. Åkermanite, anal (Humboldtite B.)	32.82	5.44	5.32	9.59	41.69	.76	3.75	99.71	H. S. Washington. Anal. Ref. same as 2. Monte Somma.
reduced	33.02	5.48	9.14	10.42	41.94				
calc.	33.55	4.63	7.84	10.70	43.28				
diff.	+0.53	-0.85	-1.30	+ .28	+1.34				
diff. by Buddington	+0.63		+ .09	+ .36	-0.69				
4. Åkermanite, anal. (Humboldtite)	34.71	3.64	6.56	12.04	40.36	.75	1.53	100.51	H. S. Washington, Ref. same as 2. Humboldtite. Monte Somma.
reduced	34.85	3.65	7.14	12.84	40.52				
calc.	35.25	3.57	7.64	12.73	40.81				
diff.	+0.40	-0.08	-0.50	-0.11	+0.29				
diff. by Buddington	+0.84		+0.18	-0.10	-0.40				

TABLE I. ANALYSES OF MELLILITES.

	CaO	Na ₂ O (K ₂ O)	MgO	Al ₂ O ₃	SiO ₂	Fe ₂ O ₃	FeO etc.	Total	REFERENCES
5. Mellilite, anal. reduced calc. diff. diff. by W. T. S.	35.41	3.24	4.15	10.30	42.07	.50	2.18	100.40	MnO, P ₂ O ₆ , CO ₂ etc. = 2.35.
	34.98	3.43	5.78	11.25	44.56				W. T. Schaller, Anal.
	33.29	4.51	6.62	11.25	44.33				Ref. Jour. Wash.
	-1.69	+1.08	+0.84	0.0	-0.23				Acad. Sci., Vol. 4, p.
	+0.26			-1.97	+1.71				473, 1914. Colorado.
6. Mellilite, anal. reduced calc. diff.	29.85	5.27	4.72	9.86	41.68	2.61	4.32	100.07	H. S. Washington, Anal. Am. J. Sci.,
		.78					H ₂ O = .98		14, 1927, p. 192.
	30.13	6.11	4.76	9.95	42.06	2.63	4.36		Villa Senni.
	30.59	5.79	4.28	9.91	42.83	2.92	3.68		
	+0.46	-0.32	-0.48	-0.04	+0.77	+0.29	-0.68		
7. Mellilite reduced calc. diff. diff. by W. T. S.	32.98	2.18	6.33	6.47	40.14	9.95	.53	100.34	H ₂ O = .27.
		1.49							F. Zambonini, ana-
	34.45	3.30	6.92	13.40	41.93				lyst. Zeit. Kryst.,
	34.63	3.48	6.78	13.38	41.73				Vol. 41, p. 266, 1906.
	+0.18	+0.18	-0.14	-0.02	-0.20				Capo di Bove.
8. Mellilite, anal. reduced calc. diff.	+0.90			-0.60	-0.30				
	33.92	3.25	6.02	10.47	41.07	3.80		99.57	Millosevich, Anal.
		1.04							Am. Jour. Sci., p.
	34.07	4.40	6.05	14.33	41.25				192, 1927. Albano.
	33.86	3.95	6.50	13.41	42.28				
	-0.21	-0.45	+0.45	-0.92	+1.03				

TABLE I. ANALYSES OF MELILITES.

	CaO	Na ₂ O (K ₂ O)	MgO	Al ₂ O ₃	SiO ₂	Fe ₂ O ₃	FeO etc.	Total	REFERENCES
9. Melilite, anal.	34.78	3.40	5.87	10.93	41.09	3.40	H ₂ O = .24	100.39	Bodlaender, Analyst. <i>Neues Jahrbuch</i> , p. 15, 1893. Capo di Bove.
reduced	35.24	.68							
calc.	34.03	3.90	5.95	13.28	41.63				
diff.	-1.21	3.95	6.49	13.76	41.77				
diff. by W. T. S.	-0.65	+0.05	+0.54	+0.48	+0.14				
10. Melilite, anal.	32.47	1.95	6.44	6.42	39.27	10.17		98.18	Damour, Analyst. See Dana-System <i>Min.</i> p. 475. Capo di Bove.
reduced	34.54	1.46							
calc.	34.85	3.09	6.85	13.75	41.77				
diff.	+0.31	3.25	6.63	13.76	41.51				
diff. by W. T. S.	+0.99	+0.16	-.22	+0.01	-.26				
11. Melilite, anal.	31.81	4.43	5.75	10.88	40.69	4.43		98.35	Damour, Analyst. Dana-Syst. <i>Min.</i> p. 475. Monte Somma.
reduced	32.92	1.36							
calc.	32.82	4.83	5.95	14.19	42.11				
diff.	-.10	4.89	6.05	14.18	42.06				
diff. by W. T. S.	+0.96	+0.06	+0.10	-.01	-.05				
12. Melilite, anal.	32.18	2.21	6.41	7.56	39.20	11.34	H ₂ O = .21	100.56	Same reference as No. 7. Capo di Bove.
reduced	33.60	1.45							
calc.	33.83	3.31	6.69	15.47	40.93				
diff.	+0.23	4.06	5.73	15.50	40.88				
diff. by W. T. S.	+1.93	+0.75	-.96	+0.03	-.05				
				-1.42	-.51				

TABLE I. ANALYSES OF MELILITES.

	CaO	Na ₂ O (K ₂ O)	MgO	Al ₂ O ₃	SiO ₂	Fe ₂ O ₃	FeO etc.	Total	REFERENCES
13. Melilite, anal.	32.05	2.12	6.71	8.61	38.34	10.02		99.36	Damour, analyst. Dana p. 475. Capo di Bove.
reduced	33.65	1.51	7.04	15.77	40.26				
calc.	33.83	4.06	5.73	15.50	40.88				
diff.	+0.18	+0.78	-1.31	-.27	+0.62				
diff. by W. T. S.	+1.93			-2.40	+0.47				
14. Gehlenite, anal.	33.19	4.18	3.12	12.89	37.84	1.13	5.66	100.33	C. E. Tilley, H ₂ O = 1.66, TiO ₂ = .66. <i>Geol.</i> <i>Mag.</i> , Vol. LXVI No. 782, p. 347-352, 1929. Co. Antrim, Ireland.
reduced	33.64	4.23	3.16	13.06	39.02	1.15	5.74		
calc.	33.95	3.71	9.35	13.88	39.11				
diff.	+0.31	-0.52	+0.45	-0.33	+0.09				
15. Gehlenite, anal. (Fuggarite)	37.65	2.04	4.89	17.97	34.04	3.49	insol = .12	100.20	E. Mayr, Analyst. <i>Zeit. Kryst.</i> , Vol. 27, p. 577, 1896. Mon- zoni.
reduced	38.10	2.06	4.94	20.45	34.45				
calc.	37.28	2.29	5.20	20.74	34.49				
diff.	-0.82	+0.23	+0.26	+0.29	+0.04				
diff. by W. T. S.	-0.62		+0.48		+0.14				
16. Gehlenite, anal.	37.90		3.88	22.02	29.78	3.22	1.63 MnO = .19	100.00	Ign. by diff. = 1.38. Rammelsberg, Ana- lyst. Dana-Syst. <i>Min.</i> , p. 476. Mon- zoni.
reduced	39.21		5.07	24.91	30.81				
calc.	40.17		4.29	24.77	30.77				
diff.	+0.96		-0.78	-0.14	-0.04				
diff. by W. T. S.	+0.95			-1.18	+0.23				

TABLE I. ANALYSES OF MELLITES.

	CaO	Na ₂ O (K ₂ O)	MgO	Al ₂ O ₃	SiO ₂	Fe ₂ O ₃	FeO etc.	Total	REFERENCES
17. Gehlenite, anal. reduced calc. diff. diff. by W. T. S.	36.74		3.77	21.33	30.01	3.56	Ign. 4.72	100.13	Lemberg, Analyst. Dana-Syst. Min., p. 476. Monzoni.
	39.03		4.01	25.08	31.88				
	39.55		3.72	24.90	31.83				
	+0.52		-0.29	-0.18	-0.05				
	+0.72			-0.84	+0.12				
18. Gehlenite, anal. reduced calc. diff.	38.11	0.33	2.20	19.80	31.60	5.97	(H ₂ O)1.53	99.54	Damour, Analyst. Dana-Syst. Min., p. 476. Monzoni.
	39.76	0.34	2.29	24.64	32.97				
	38.53	0.35	3.14	24.85	33.13				
	-1.23	+0.01	+0.85	+0.21	+0.16				
19. Gehlenite, anal. (velardénite) reduced calc. diff.	40.86		4.18	25.52	27.88	1.59	0.43	100.80	E. V. Shannon, Ana- lyst. Proc. U. S. National Museum, Vol. 60, Art. 22, p. 1-4. H ₂ O=0.34. Tulare Co., Calif.
	40.67		4.17	25.40	27.75	1.58			
	40.91		4.26	26.46	28.37				
	+0.24		-.34	-0.52	+0.62				
20. Gehlenite, anal. (Velardénite) reduced calc. diff. diff. by W. T. S.	39.55	0.31	2.44	27.82	26.33	1.43	0.51	100.27	H ₂ O=1.85 Allen, Analyst. Am. Jour. Sci., Vol. 5, p. 527, 1908. Velar- dena.
	40.19	0.31	2.99	29.72	26.79				
	40.53	0.23	2.94	29.46	26.84				
	+0.34	-0.08	-0.05	-0.26	+0.05				
	-.33			+0.22	+0.11				

TABLE I. ANALYSES OF MELILITES.

	CaO	Na ₂ O (K ₂ O)	MgO	Al ₂ O ₃	SiO ₂	Fe ₂ O ₃	FeO etc.	Total	REFERENCES
21. Sarcolite, anal. reduced calc. diff. diff. by W. T. S.	33.70	4.43	0.36	21.63	39.34			99.46	A. Pauly, analyst. <i>Centr. Min.</i> , p. 266, 1906. Monte Somma.
	33.88	4.45	.36	21.75	39.56				
	31.96	4.12	1.53	22.31	40.08				
	-1.92	-.33	+1.17	+0.56	+0.52				
	-.74			+0.21	+0.53				
22. Sarcolite, anal. reduced calc. diff. diff. by W. T. S.	32.36	3.30		21.54	40.51			98.91	Rammelsberg, Analyst. <i>Pogg. Ann.</i> , Vol. 109, p. 570, 1860. Monte Somma.
		1.20							
	32.85	4.16		21.87	41.12				
	31.23	3.57		23.54	41.66				
	-1.62	-0.59		+1.67	+0.54				
	+0.51			+0.69	-1.20				
23. Sarcolite, anal. reduced calc. diff. diff. by W. T. S.	32.43	2.93		24.50	42.11			101.97	Scacchi, Analyst. <i>Dana-Syst. Min.</i> , 5th ed., p. 318, 1842. Monte Somma.
	31.80	2.88		24.03	41.29				
	32.06	2.86		23.91	41.17				
	+0.26	-0.02		-0.12	-0.12				
	+2.78			-1.45	-1.33				

TABLE II.
ATOMIC COMPOSITION OF THE MEILLITES

Anal. No.	Calculated							Found							Ca +Na	Mg+Al +Si
	Mineral name	Ca	Na	Mg	Al	Si	O	Ca	Na	Mg	Al	Si	O			
1	Åkermanite	19.1		8.9	.4	20.7	70	18.8		8.9	0.5	20.7	70	18.8	30.1	
2	Åkermanite (Humboldtite)	17.6	2.4	5.8	6.0	18.2	70	17.3	2.4	5.9	6.1	18.1	70	19.5	30.1	
3	Åkermanite (Humboldtite)	16.0	4.0	5.2	5.6	19.2	70	16.2	4.6	5.1	5.4	19.1	70	20.8	29.6	
4	Åkermanite (Humboldtite)	16.9	3.1	5.1	6.7	18.2	70	16.9	3.1	5.1	6.7	18.2	70	20.0	30.0	
5	Meliite	15.9	2.9	4.4	5.9	19.7	70	16.6	2.9	3.8	5.9	19.7	70	19.5	29.4	
6	Meliite	14.9	5.1	4.3	6.3	19.4	70	14.8	5.2	4.9	6.2	19.2	70	20.	30.3	
7	Meliite	16.4	2.8	4.5	7.0	18.5	70	16.5	3.0	4.5	7.0	18.5	70	19.5	30.0	
8	Meliite	16.1	3.4	4.3	7.0	18.7	70	16.5	3.4	4.1	6.9	18.6	70	19.9	29.6	
9	Meliite	16.2	3.4	4.3	7.2	18.5	70	16.9	3.4	4.0	7.0	18.5	70	20.3	29.5	
10	Meliite	16.6	2.8	4.4	7.2	18.4	70	16.4	2.7	4.5	7.2	18.4	70	19.1	30.1	
11	Meliite	15.6	4.2	4.0	7.2	18.6	70	15.6	4.2	4.0	7.4	18.6	70	19.8	30.0	
12	Meliite	16.1	3.5	3.8	8.1	18.1	70	15.9	2.8	4.4	8.1	18.1	70	18.7	30.6	
13	Meliite	16.1	3.5	3.8	8.1	18.1	70	16.0	2.8	4.7	8.2	17.8	70	18.8	30.7	
14	Gehlenite	16.7	3.3	4.6	7.5	17.9	70	16.7	3.7	4.4	7.5	17.9	70	20.4	29.8	
15	Gehlenite (Fuggarite)	18.0	2.0	3.5	11.0	15.5	70	18.4	1.8	3.4	10.9	15.5	70	20.2	29.8	
16	Gehlenite	19.5		2.9	13.2	13.9	70	19.0		3.4	13.2	13.9	70	19.0	30.5	
17	Gehlenite	19.1		2.5	13.2	14.3	70	18.8		2.7	13.2	14.3	70	18.8	30.2	
18	Gehlenite	18.5	0.3	2.1	13.1	14.8	70	19.0	0.3	1.5	13.1	14.8	70	19.3	29.4	
19	Gehlenite	20.0		2.9	14.2	12.9	70	20.0		3.0	14.3	12.7	70	20.	30.0	
20	Gehlenite (Velardénite)	19.8	0.2	2.0	15.8	12.2	70	19.8	0.2	1.9	15.7	12.2	70	20.	29.8	
21	Sarcosite	15.0	3.5	1.0	11.5	17.5	70	16.0	3.8	0.2	11.3	17.4	70	19.8	28.9	
22	Sarcosite	14.5	3.0		12.0	18.0	70	15.4	3.5		11.3	18.0	70	18.9	29.3	
23	Sarcosite	14.9	2.4		12.2	17.8	70	14.8	2.4		12.2	17.8	70	17.2	30.0	

TABLE III
MOLECULAR PERCENTAGE COMPOSITION OF THE MELILITE GROUP

A. $\text{Ca}_2\text{Al}_2\text{SiO}_7$ Gehlenite molecule. B. $\text{Ca}_2\text{MgSi}_2\text{O}_7$ Åkermanite molecule. C. $\text{Na}_2\text{Si}_3\text{O}_7$ Soda-melilite molecule. D. CaSi_3O_7 Sub-melilite molecule.						
No.	Anal. Mineral	A	B	C	D	Schaller's anal. no.
1.	Åkermanite	2.0	90.7	0.0	7.3	4 and 5
2.	Åkermanite (Humboldtite)	30.6	58.6	10.8	0.0	
3.	Åkermanite (Humboldtite)	28.7	53.1	18.2	0.0	
4.	Åkermanite (Humboldtite)	34.3	51.7	14.0	0.0	
5.	Melilite	30.9	45.8	13.4	9.9	16
6.	Melilite	32.8	45.1	22.1	0.0	
7.	Melilite	36.1	46.1	13.7	4.1	13
8.	Melilite	36.2	44.2	15.5	4.1	
9.	Melilite	37.1	44.1	15.5	3.3	11
10.	Melilite	37.2	45.2	12.8	4.8	12
11.	Melilite	38.7	41.6	18.0	1.7	10
12.	Melilite	41.8	38.9	15.9	3.4	15
13.	Melilite	41.8	38.9	15.9	3.4	14
14.	Gehlenite	50.9	37.2	11.9	0.0	
15.	Gehlenite (Fuggarite)	55.7	35.3	9.0	0.0	17
16.	Gehlenite	66.8	29.2	0.0	4.0	7
17.	Gehlenite	67.3	25.4	0.0	7.3	8
18.	Gehlenite	67.3	21.5	1.4	9.8	9
19.	Gehlenite	71.1	28.9	0.0	0.0	
20.	Gehlenite (Velardeñite)	79.2	19.9	0.9	0.0	6
21.	Sarcolite	62.8	10.4	18.5	8.3	3
22.	Sarcolite	64.5	0.0	14.3	21.2	2
23.	Sarcolite	65.6	0.0	11.4	23.0	1

However the expected total of $\text{Ca} + \text{Na} = 20$ is not in evidence for most analyses although the assumed end members have each 20 atoms of calcium. There is the significant fact evident, however, that the total of $\text{Ca} + \text{Na}$ does not materially exceed 20 atoms in any analysis. This deficiency it seems cannot be explained by error

in analyses and must be accounted for in an adequate explanation of the series. A general formula of the melilites may from these deductions be written for the whole group, as follows:



From the analyses as given, the probable limits of x , y , z , can be stated as follows:

(a) x may be 0 or some small number probably not greater than 3 (in any known melilite).

(b) y may have as its maximum value 10 and its minimum value 0. This corresponds in the first case to a melilite composed entirely of the end member âkermanite, and where $\text{Mg} = 0$ to a melilite with no âkermanite present; it is evident that a value greater than 10 Mg would necessitate more than 20 atoms in the $\text{Ca} + \text{Na}$ member, and this is not found in the series.

(c) z varies from 0 to 15.7 in the series. This variation indicates that the gehlenite molecule ($\text{Ca}_2\text{Al}_2\text{SiO}_7$) is absent in some melilites and that velardeñite (#20) is an almost pure gehlenite, the pure molecule having 20 atoms. The value 20 may be taken as the maximum Al content of the melilites since more aluminum would necessitate more than 20 $\text{Ca} + \text{Na}$ atoms.

(d) The silicon as given in the general formula is $30 - z$ atoms. Inspection of the table shows that the minimum number of silicon atoms is ten. The maximum number of atoms noted in the table is 20.7. If however y and z could both be zero then the maximum number would have to be 30. In Winchell's hypothetical soda melilite this value is used and the formula is $\text{Na}_{20}\text{Si}_{30}\text{O}_{70}$. This molecule will be discussed in another section of this paper. There is another possibility of using the theoretically maximum amount of silicon in a formula such as $\text{Ca}_{10}\text{Si}_{30}\text{O}_{70}$. This formula has only 10 Ca atoms rather than 20 (or a number near 20) and therefore cannot be considered as a true melilite molecule. However, it can be used to explain the fact that the total $\text{Ca} + \text{Na}$ is in most cases less than 20, as will be shown later.

An examination of the group formula indicates that sodium replaces calcium and that Mg, Al, Si are replaceable to the extent noted above. In the tables of ionic radii of Goldschmidt,⁸ which are now generally accepted, the calcium and sodium ions are of the

same order of magnitude, the Mg, Al, Si are considerably smaller and of comparative similar radii among themselves. Oxygen is the largest of the atoms present in the composition of the series. There is then an adequate reason for the relationship established. There are seemingly two kinds of basic elements, the Ca and Na together, and the Mg, Al, Si elements in another set. Thus Winchell's R_5O_7 is really $X_2Y_3O_7$ which is the less exact form of the author's general formula as given above.

The question as to the replaceability of calcium and sodium atom for atom cannot be definitely answered by the data in this paper because there is no real constancy in the number of atoms of both in the melilites. However, in other recent treatments of the relationship of these two atoms in isomorphous compounds there is little doubt left but that these two elements can substitute for each other, atom for atom, provided the valences are balanced after substitution by some further change such as for example an attendant substitution of magnesium for aluminum. The amphiboles, pyroxenes, feldspars,⁹ etc. are evidences of this type of replacement.

As to the replacement of Mg, Al, and Si there seems to be excellent evidence in the actual analyses of this series that this has taken place. It is gratifying to the author that while this paper was in preparation, that further evidences of this type of replacement were pointed out in tourmaline¹⁰ and the amphiboles and pyroxenes¹¹ by F. Machatschki in two very recent papers. The problem of composition and isomorphism is treated in a manner similar to that given in this paper.

That the atomic volume relationships are not the only determining factors in isomorphism is obvious. What the other factors may be are not as yet known, but it is safe to say that volume isomorphism is one of the most important considerations in the study of a series such as the melilites, or, as Machatschki has pointed out, in the tourmalines and amphiboles and probably all other silicates.

The end members adopted by the author have been chosen to agree with the general formula as follows:

- | | |
|--------------------|-----------------------|
| A. $Ca_2Al_2SiO_7$ | Gehlenite |
| B. $Ca_2MgSi_2O_7$ | Åkermanite |
| C. $Na_2Si_3O_7$ | Soda melilite |
| D. $CaSi_3O_7$ | Sub-melilite molecule |

The molecules A and B can enter into combination in all proportions as indicated by the analyses of natural melilites and by the work of Ferguson and Buddington on synthetic preparations. $\text{Na}_2\text{Si}_3\text{O}_7$ enters only to a limited extent as indicated in the analyses. This hypothetical molecule does not occur in the natural melilites to a greater extent than about 23 per cent.

CaSi_3O_7 , the sub-melilite molecule can be used to show the extent to which the formula falls short of having $(\text{Ca} + \text{Na}) = 20$. This molecule is usually of very minor importance in the composition exceeding 10 per cent in only two analyses. This type of molecule is interpreted by the author as being an indication that part of the melilite structure is open, because all the negative charges of oxygen have already been satisfied by $\text{Mg} + \text{Al} + \text{Si}$, which is constant and more firmly bound in the structure, and ionic equilibrium, so to speak, has been reached without filling up the holes which normally would be occupied by Ca or Na. That this can occur to only a limited extent without the breakdown of the structure is evident, so that such a molecule could only be expected to be present in limited amounts. CaSi_3O_7 need not be of the same structural type as the other members of the group, and still enter in the solid solution to the limited extent noted in the analyses of the natural occurrences. It is doubtful whether CaSi_3O_7 , prepared artificially, would be a true melilite.

II. Other THEORIES

The principal theories advanced to explain the composition of the melilite group have been those of Schaller, Buddington and Winchell.

Schaller proposed as end members the following:

Sarcolite	$3\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 3\text{SiO}_2$ or $\text{Ca}_3\text{Al}_2\text{Si}_3\text{O}_{12}$
Soda-sarcolite	$3\text{Na}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 3\text{SiO}_2$ $\text{Na}_6\text{Al}_2\text{Si}_3\text{O}_{12}$
Åkermanite	$4\text{MgO} \cdot 8\text{CaO} \cdot 9\text{SiO}_2$ $\text{Mg}_8\text{Mg}_4\text{Si}_9\text{O}_{30}$
Velardeñite	$2\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot \text{SiO}_2$ $\text{Ca}_2\text{Al}_2\text{SiO}_7$

Winchell has pointed out that the compounds proposed by Schaller are much dissimilar in character and not likely to be end members of the same group. In general Schaller's end members give calculated compositions about as well as those of the author. Soda sarcolite, in small amounts combined with a large amount of sar-

colite gives agreement with the end members $\text{Ca}_2\text{Al}_2\text{SiO}_7$, $\text{Na}_2\text{Si}_3\text{O}_7$ and CaSi_3O_7 proposed by the author. The other two end members are essentially the same as the author's molecules. An objection which may be raised against Schaller's molecules is that sarcolite ($\text{Ca}_3\text{Al}_2\text{Si}_3\text{O}_{12}$) and velardeñite ($\text{Ca}_2\text{Al}_2\text{SiO}_7$) are composed of the same elements in slightly differing amounts. That the melilites can have two end members of such similarity seems unlikely.

The chief difference between the end members proposed by Winchell and those of the author is that Winchell has as one of his compounds $\text{Ca}_3\text{Si}_2\text{O}_7$. An examination of this molecule in comparison with the type formula of the author suggests that it is not likely to yield satisfactory results because the 3:2:7 ratio is not found to be present in the natural melilites. When this molecule is used there is an apparent excess of silica in the analyses which may be as high as 9 per cent. The molecule used by the author can be derived from $\text{Ca}_3\text{Si}_2\text{O}_7$ and silica in the following manner:



CaSi_3O_7 can therefore be used and no excess of SiO_2 will be observed in the analysis. The excess silica is explained by Winchell as being interatomic because the silicon and oxygen atoms are small enough to fit into the atoms without disturbing them. At the time that his paper was written there had not been much experimental work to verify the size of the oxygen atoms and it was considered one of the smallest. Present measurements, however, are in general agreement that oxygen is one of the largest of atoms entering into the composition of the silicates, so that this theory seems highly improbable on the basis of later knowledge.

Buddington has attempted to show that the end members proposed by Schaller form solid solutions, by preparing synthetic mixtures corresponding to the end members and studying the properties of the resultant compounds prepared under the proper conditions. As Winchell has pointed out, his evidence is rather inconclusive. There are many cases where nonhomogeneous crystallizations result. In fact, when the sarcolite molecule, proposed by Schaller, was used with artificial åkermanite and gehlenite, the whole series failed to show solid solution. Only when a

charge approximating the actual sarcolite composition was used did homogeneous crystallizations result. This seems, to the author, to indicate that the sarcolite and soda sarcolite molecules are not an adequate explanation of the composition of natural sarcolite. In this paper no attempt has been made to apply the molecules proposed to an analysis of Buddington's synthetic mixtures since some of the components used are, in the author's opinion, not those which occur in the natural melilites and, therefore, out of the scope of this paper, which is concerned only with the natural occurrences.

DISCUSSION OF ANALYSES

All the available satisfactory analyses of members of the melilite group are given in Table I. The calculated values given in the table are derived from the theoretical composition as computed from the author's molecules. In reducing the analyses Al_2O_3 and Fe_2O_3 are grouped together, as are Na_2O and K_2O , and also FeO , MgO and MnO . Only where the FeO or Fe_2O_3 are important constituents are they calculated separately as iron-åkermanite or iron-gehlenite. Differences between calculated and reduced analyses are given in order to quickly show the divergence of the theoretical composition from the actual analysis. Differences obtained by use of Schaller's molecules are also given for comparison. The numbers of the analyses in Table I are used in Tables II and III for the corresponding atomic and molecular compositions, respectively.

Since there are already adequate names in established use for the various members of the group no new ones will be here proposed. The various members of the group are here defined in terms of their molecular percentages as follows:

(a) Åkermanite—those members of the group giving over 50 per cent of the åkermanite molecule ($\text{Ca}_2\text{MgSi}_2\text{O}_7$) and less than 25 per cent of the soda-melilite and sub-melilite molecules combined ($\text{Na}_2\text{Si}_3\text{O}_7 + \text{CaSi}_3\text{O}_7$).

(b) Gehlenite—those members of the group having over 50 per cent of the gehlenite molecule ($\text{Ca}_2\text{Al}_2\text{SiO}_7$) and less than 25 per cent of the soda-melilite and sub-melilite molecules combined ($\text{Na}_2\text{Si}_3\text{O}_7 + \text{CaSi}_3\text{O}_7$).

(c) Melilite—those members of the group which have neither the åkermanite molecule nor the gehlenite molecule present to the extent of 50 per cent, and have less than 25 per cent of the other two molecules combined.

(d) Sarcolite—those members of the group having over 25 per cent of the soda-melilite and sub-melilite molecules combined.

The above classification is in conformity with the long established nomenclature in the group. The more recent names, velardénite and humboldtilite, are not used. Fuggarite is here considered as a gehlenite. Since this name has been applied to but one member of the group which is within the limits defined for gehlenite there seems to be no reason for retaining the name in the classification.

ÅKERMANITE

There are two analyses available of almost pure åkermanite (Anal. 1,—Table I) which are obviously of the same material. The calculated composition agrees very well with the observed percentages. Analyses 2, 3, 4, of Table I (Humboldtilite of Buddington) are also good analyses and agree with the calculated compositions. There is a considerably greater percentage of the gehlenite molecule in these three åkermanites than in the other member noted above.

MELILITE

The analyses of these intermediate members of the group are in general agreement with the theory. The substitution of Al_2O_3 for Fe_2O_3 is not accompanied by any appreciable discrepancy in the calculated composition (see analyses 12, 13). There are some analyses for which the theory does not hold very accurately but in view of the fact that most of the analyses do adequately fit the end members, it seems likely that obvious discrepancies may be explained by impurities in analyzed material, and other sources of error. Table III, giving the molecular composition, shows that these members have about 16 per cent of the soda melilite molecule and about 4 per cent of the sub-melilite molecule.

GEHLENITE

There are two modern analyses of this species both of which agree well with the theoretical composition (Anal. 19 and 20). The

others, while older analyses, are in rather good agreement also, with the exception of analysis 18, which has a deficiency of 1.23 per cent of CaO. The gehlenites in general have a low percentage of the minor molecules $\text{Na}_2\text{Si}_3\text{O}_7$ and CaSi_3O_7 .

SARCOLITE

The sarcolites differ from the other members of the group in having a rather large percentage of the soda melilite and sub-melilite molecules. The calculated compositions do not agree especially well with the analyses. The two older analyses are not especially good ones (Anal. 22 and 23, Table I). The more recent one by Pauly, however, does not give good agreement with the theory. The calculated compositions by Schaller are also not in very close agreement with the observed percentages.

The fact that this species has such a large percentage of the sub-melilite molecule as compared to the other members of the group leads to a speculation as to whether it is really a true melilite. An x-ray spectrographic study of sarcolite with this in view will be undertaken later as well as a new analysis. Gossner¹² in a recent paper has shown that sarcolite is not closely related to the melilites but instead is related to the scapolites.

SUMMARY AND CONCLUSIONS

The theory as to the composition of the melilites may be here summed up as follows:

General formula:



$$\begin{aligned} \text{where } x &= 0 \text{ to } 3 \\ y &= 0 \text{ to } 10 \\ z &= 0 \text{ to } 20 \end{aligned}$$

This formula gives rise to the melilite molecules as follows:

Åkermanite	$\text{Ca}_2\text{MgSi}_2\text{O}_7$
Gehlenite	$\text{Ca}_2\text{Al}_2\text{SiO}_7$
Soda melilite	$\text{Na}_2\text{Si}_3\text{O}_7$
Sub-melilite	CaSi_3O_7

The first two of these may be present in any amount. The third (soda-melilite) in the natural occurrences does not exceed

25 per cent, the fourth (sub-melilite molecule) is the least important and with but two exceptions does not exceed 10 per cent.

There are no facilities in this laboratory to conduct experiments on the artificial compounds of the compositions of these end members, it is suggested that this would possibly prove an interesting problem. The study of the space group of melilite and a comparison of the x-ray spectrographs of gehlenite, melilite and sarcolite will be published at a later date.

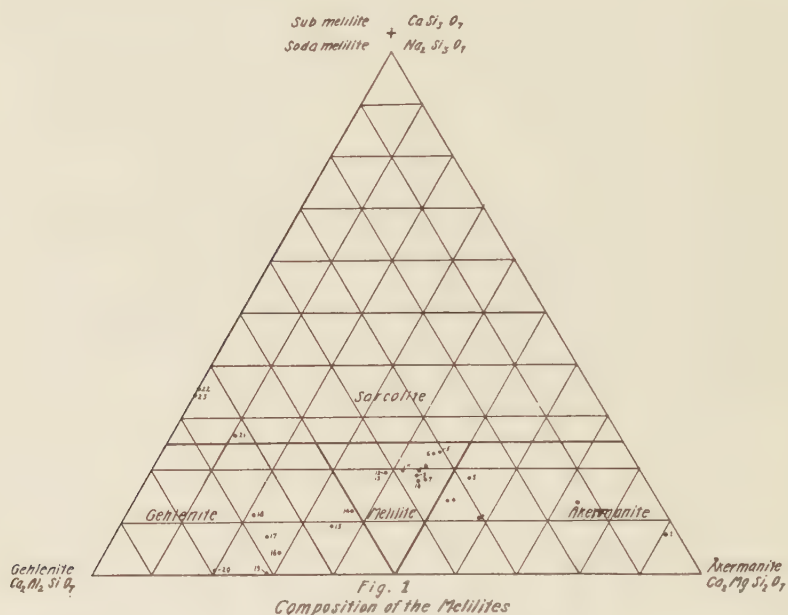


Fig. 1
Composition of the Melilites

The optical properties of the various members of the group have not been recorded in this paper because in the artificial $\text{Ca}_2\text{Al}_2\text{SiO}_7$ — $\text{Ca}_2\text{MgSi}_2\text{O}_7$ series a careful optical study was made.

Since these are the two dominant molecules present the principal optical differences arise from the percentages of these present. An additional reason for omitting an optical study was lack of type material in order to study the effect of the soda melilite and sub-melilite molecules on the optical properties.

A diagram (Fig. 1) shows the composition of the various members of the melilite group. The soda melilite molecule and sub-

melilite molecule have been combined in order to better represent the composition on a plane diagram.

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THE STRONTIUM OCCURRENCE NEAR LA CONNER, WASHINGTON

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A strontium deposit from which celestite and small amounts of strontianite were mined during the latter part of the war is located at the southeast corner of Fidalgo Island in western Skagit County. The nearest town is LaConner, situated about one mile to the northeast and on the mainland. Fidalgo Island lies southeast of the main San Juan group in north Puget Sound and is separated from the mainland by a very narrow pass known as Swinomish Slough, through which the tide moves with considerable velocity. To reach the strontium deposit it is necessary to go by boat from LaConner through the south end of the slough to the open sound and then a short distance westward along the southward facing cliffs of Fidalgo Island. The deposit is marked by an abandoned mine tunnel situated a few feet above tide level.

Fidalgo Island is made up largely of massive rocks and has a very uneven topography. The land lying east of Fidalgo is, on the contrary, very flat, and is a part of the extensive Skagit River delta. Here and there the level surface of the delta is interrupted by rocky and precipitous hills which once were islands, similar to but smaller than Fidalgo and which have become tied to the mainland by the outward extension of the river flood plain.

The geology of Fidalgo Island has never been studied in detail. R. D. McLellan,¹ in the map accompanying his report on the geology of the San Juan Islands, covers only portions of the western side of Fidalgo Island. The country rock in the vicinity of the strontium deposit probably belongs to the Fidalgo formation, referred by McLellan to the Triassic.² The following description is quoted from McLellan's report:³

"The Fidalgo formation is composed of three distinct rock types which are invariably associated with each other: (1) Large irregular masses of extremely coarse-grained dunite, which weathers to a dark green or dark brown color. (2) Thin irregular off-shoots or fine-grained dunite, injected into the joint-cracks of the coarse-

¹ R. D. McLellan, The geology of the San Juan Islands. *Univ. of Wash. publications in geology*, vol. 2, Nov. 1927.

² *Op. cit.*, p. 144.

³ *Op. cit.*, pp. 142 and 144.

grained variety. These off-shoots weather first to a light brown or buff color, and on further disintegration, to a bright orange-red. (3) Both types of dunite are everywhere cut by thin stringers (rarely exceeding two inches in thickness) of serpentinized pyroxenite. The latter is composed almost entirely of altered diallage, the crystals often being as large as the width of the stringer permits. Of the three rock types the pyroxenite is the least resistant to alteration and erosion."

A specimen collected by the writer from a point 50 feet west of the celestite vein outcrop consisted mainly of olivine and serpentine with subordinate calcite and epidote. Of a later age are large amounts of quartz and pyrite. The quartz occurs both in veins and in scattered but fairly pure masses less than an inch across and sometimes with a vuggy tendency. The pyrite is impregnated through the rock in scattered small grains. It is found with or without the quartz, in fact the latter occurrence is the commoner. The country rock specimens collected immediately adjacent to the deposit were not pyritized, but were calcitized instead.

Although the LaConner strontium deposit has been mentioned a few times in print,⁴ no detailed description of it has as yet been published. This locality was visited by the writer in 1921 and again in 1928. The deposit consists mainly of celestite which occupies a shattered zone running north 65 degrees east and dipping 42 degrees to the northwest. The lode width varies between 3 and 4 feet, but only rarely does that entire distance contain vein minerals. The latter occur in stringers and pods which parallel the strike of the shattered zone. Sometimes the country rock predominates in this zone with the celestite and associated minerals subordinate.

The deposit crops out from tide level to the top of the hill and then can be traced northeastward for a short distance until obscured by the vegetation. About eight feet above high tide a 110 foot tunnel has been driven on the vein. At the top of the hill a trench about 100 feet long and 2 to 10 feet deep into the vein has been excavated. The bottom of the trench lies about 80 feet above the tunnel level. The vein at this point is about 30 inches wide and

⁴ Hill, J. M., *U. S. Geol. Survey, Mineral Resources*, 1915, pt. 2, p. 187, and 1916, pt. 2, p. 194. Patty and Glover, *Wash. Geol. Survey, Mineral Resources of Washington*, Bull. 21, p. 113, 1919. Shedd, Solon, *Wash. Geol. Survey, Bull.* 30, pp. 144-145, 1922.

very rich. This development took place during the war. The small tonnage of strontium minerals secured was shipped by barge to Seattle.

Celestite is quantitatively the most important of the vein minerals, but there is also considerable strontianite. Calcite, dolomite, and limonite are present, but the last two named are extremely subordinate. Descriptions of the individual minerals follow:

CELESTITE. The characteristic bluish tinge of this mineral can be discerned in most of the specimens collected. The depth of the blue varies considerably. In some instances the celestite is either milky or clear and colorless. The characteristic basal cleavage is very well developed, but the prismatic cleavage is less apparent in the unaltered mineral. However, most of the celestite is altered in part at least to strontianite and the tendency of the replacing mineral to follow cleavage directions in the form of minute veins causes the prismatic cleavage to show up very plainly on the basal cleavage face. The strontianite is whiter in color and of denser appearance. In some specimens the alteration to strontianite is practically complete, the only evidence of the former presence of celestite being a "ghost" cleavage. No euhedral or even anhedral crystals were found. However, partial dissolution by salt water of celestite-bearing rocks along the beach has brought about the formation of some peculiar shapes. Dissolution proceeded with greatest ease along the planes of the macro and brachydomes, producing an eight-faced form. This is not pseudo-octahedral, however, because the dissolution of one dome is very much greater than the other. These water-worn celestite specimens are milky-white and translucent. The surfaces of the dome faces exhibit a sort of pillow structure on a very fine scale. The dissolving power of salt water on celestite has already been pointed out by Kraus.⁵ The Fidalgo Island celestite is the only primary mineral of importance in the vein.

CALCITE. That considerable calcium carbonate was present in the celestite-bearing solutions is evidenced by the extreme calcitization of "horses" of country rock in the shattered zone and by the occasional presence in the deposit of veins of pure calcite. The calcite is white in color and coarsely crystalline. The crystals are arranged normal to the walls of the veins which are rarely over an

⁵ Kraus, E. H., The occurrence and distribution of celestite-bearing rocks. *Amer. Jour. of Science*, 4th ser., vol. 19, pp. 286-293, 1905.

inch in thickness. No specimens were obtained containing both calcite and celestite, so the age relationship between these two minerals could not be determined. A very small amount of dolomite occurs with the calcite.

STRONTIANITE. This mineral is obviously of secondary deposition. It occurs as an alteration product of both celestite and calcite. In the former instance the strontianite coats the surface of the celestite and is also found within the latter mineral in veins along cleavage cracks. These vary in thickness from a size visible only under the microscope to a stage where alteration is virtually complete, and only the celestite cleavage remains as a clue to the identity of the mineral formerly occupying that space. Strontianite coats the surface of the calcite or lines small vugs within, but does not penetrate through the calcite in veins to the degree it does with celestite. The strontianite is white where pure and the crystallization is so fine as to give the material a powdery appearance. The most striking characteristic of the Fidalgo Island strontianite is its habit. On the surface it generally appears in botryoidal or nodular form, while within the specimen the mineral is characteristically cellular or reticulated. At times the strontianite is yellow or brown in color due to the presence of limonite. The obviously later age of the strontianite plus its habit lead the writer to believe that it is a secondary mineral and was formed by the action of carbon dioxide bearing ground water on the vein celestite. Furthermore, it has been noted "that the principal bodies of the carbonate are found within 10 feet of the surface and are more abundant where the vein is capped by trees and moss, which suggests that organic acids may have been responsible for the alteration."⁶ Sherzer has noted the alteration of celestite to strontianite in the Plum Creek quarries, Michigan.⁷

LIMONITE. This mineral is found in very minor amounts associated with the strontianite. The iron was undoubtedly derived from the serpentine country rock.

The Fidalgo Island strontium minerals are rather unique in their occurrence. Celestite and strontianite have been described from a number of other localities in the United States such as Cal-

⁶ Hill, J. M., *U. S. Geol. Survey, Min. Resources*, 1916, pt. 2, p. 187.

⁷ Sherzer, W. A., *Am. Jour. Sci.*, 3rd series, vol. 50, p. 246, 1895.

ifornia, Arizona, Texas, Ohio, New York and West Virginia.⁸ In all of these occurrences, however, the strontium minerals are found in sedimentary rocks, usually limestone and dolomite, but sometimes with salt, gypsum, and clay. The strontium is considered by most investigators to have been deposited simultaneously with the other sedimentary material. Later ground water activity caused a concentration of the strontium (generally in the form of celestite) along water courses. In only one locality was the presence of igneous rock mentioned. That was in Arizona where igneous flows and dikes are numerous through the sedimentary series.⁹ The same general associations apply to the foreign deposits of strontium minerals. Celestite in the Bristol district in England occurs in marl, while in Sicily it is associated with sulphur and gypsum. Strontianite occurs in commercial deposits in Westphalia, Prussia. The country rock at this locality is Cretaceous marl and limestone.¹⁰

However, strontium minerals have also been reported in hydrothermal deposits. Hill notes that celestite is a gangue mineral at the Lead Hill mines, Salina, Utah.¹¹ Strontianite occurs in England only in mineral veins, as a rarity.¹² In the Fitzroy township, Ontario, considerable celestite occurs in galena-bearing veins.¹³ In describing a deposit in Carlton County, Ontario, Spence states: "Vein has . . . originated upon a line of structural dislocation

⁸ Pratt, J. H., Strontium ores: *Mineral Resources*, 1901, pp. 1905-1958. Phalen, W. C., Celestite deposits of California and Arizona: *U. S. Geol. Survey, Bull.* 540, pp. 521-533, 1914. Culin, F. L., Celestite and Strontianite: *Ariz. Univ. Bureau of Mines, Bull.* 35, p. 4, 1916. Kraus, E. H., Occurrence of celestite near Syracuse New York: *Am. Jour. Sci.*, 4th ser., vol. 18, pp. 30-39, 1904; Occurrence and distribution of celestite-bearing rocks: *Am. Jour. Sci.*, 4th ser., vol. 19, pp. 286-293, 1905. Kraus and Hunt, The occurrence of sulphur and celestite at Maybee, Mich.: *Am. Jour. Sci.*, 4th ser., vol. 21, p. 237, 1906. Hawkins, A. C., Notes on pyrite and celestite from Rochester, New York: *Am. Mineralogist*, vol. 11, p. 165, 1926. Hess, F. L., Texas celestite deposits: *Eng. and Mining Jour.*, vol. 88, p. 117, 1909. Knopf, A., Strontianite deposits near Barstow, Calif., *U. S. Geol. Survey, Bull.* 655, pp. 267-270, 1918. Hill, J. M., Strontium ores and salts: *U. S. Geol. Survey, Mineral Resources*, 1912, p. 960.

⁹ Culin, F. L., *loc. cit.*

¹⁰ Spence, H. S., Barium deposits of Canada: *Canada Mines Rept.*, 1922, p. 100.

¹¹ Hill, J. M.: *U. S. Geol. Survey, Mineral Resources*, 1915, p. 186.

¹² Sherlock, R. L., Celestine and strontianite: *Mem. Geol. Survey, Special reports on the mineral resources Great Britain*, vol. 3, p. 51, 1918.

¹³ Young, G. A., Geology and economic minerals of Canada: *Canadian Geol. Survey*, vol. 2065, p. 54, 1926.

in the limestone. . . . The celestite has probably been deposited by ascending solutions from a deep-seated source, rather than concentrated from the enclosing crystalline limestone."¹⁴ Small amounts of sphalerite and chalcopyrite as minute crystals on calcite are noted associated with this celestite. The same writer describes the occurrence of celestite in irregular masses in brown dolomite in Renfrew County, Ontario.¹⁵ He believes that the dolomite was originally a crystalline limestone (which is the normal country rock) and was altered by the same ascending solutions which introduced the celestite. A diabase dike forms the foot wall to the deposit. The magnesium and strontium bearing solutions are thought by Spence to have been brought in by the diabase.

The writer believes that the Fidalgo Island celestite is of hydrothermal rather than ground water origin. Reasons for this belief follow: (1) The complete absence of limestone or dolomite or other sedimentary rocks in which the primary strontium could have been deposited. (2) The presence in the locality of large amounts of igneous rock. (3) The confinement of the celestite to a very narrow steeply inclined zone in the rock. (4) The presence of impregnated pyrite and quartz veins in the near-by country rock.

The stages involved in the formation of this deposit were thought to have been as follows: (1) The intrusion of the dunite. (2) The intrusion and solidification of a younger magma not yet exposed at the surface in this immediate vicinity. McLellan¹⁶ describes considerable late Jurassic igneous intrusive activity throughout this area. (3) The upward and outward movement of solutions from the consolidated magna which caused first the deposition of quartz and pyrite in the dunite as already described, and later the precipitation of celestite and calcite along a zone of weakness in the dunite. (4) Ground-water activity which caused the alteration of some of the celestite and calcite to strontianite.

¹⁴ *Op. cit.*, p. 78.

¹⁵ *Op. cit.*, p. 81.

¹⁶ *Op. cit.*

THE DOUBLE VARIATION METHOD OF REFRACTIVE INDEX DETERMINATION¹

(SECOND PAPER)

R. C. EMMONS, *University of Wisconsin.*

In the preliminary paper recently published² a method was described for determining the refractive indices of crystalline substances under the petrographic microscope by using the combination of variable temperature and variable wave length of light. The advantages pointed out are much greater speed, convenience, and accuracy. The results obtained yield also a figure for the dispersion of the substance.³ I shall review briefly the essential points of the earlier paper.

SUMMARY OF PRELIMINARY PAPER.

Crystal identification by refractive index with the petrographic microscope in white light and under ordinary working conditions has well known disadvantages. Results are most commonly stated with the modification $\pm .003$ or $\pm .005$; and these results are usually secured at the cost of considerable time and painful effort. A much greater degree of accuracy is obtained by using monochromatic light as many workers have done for years. A few have sped up their determinations by employing variable temperature of the immersion medium thereby avoiding the necessity of mixing media of different index which is quite tedious. For this purpose the electrically heated stage was commonly used. It was noticed that by combining these two procedures a peculiar

¹ Several workers in this field have by personal communication from rather widely scattered points indicated their interest in the preliminary paper. I have received many friendly criticisms and suggestions which I readily acknowledge. It is intended to include in this paper replies to the points brought up in the correspondence. Dr. H. E. Merwin of the Geophysical Laboratory very kindly pointed out my incorrect use of the term dispersion as applied to the change of index with change of temperature and I have therefore altered the title from Double Dispersion to the present one.

² *American Mineralogist*, vol. 13, 1928, p. 504.

³ This was suggested as a critical determinative factor to Dr. A. N. Winchell who has concurred and has included a table of the dispersion of minerals in volume 3 of his Optical Mineralogy, now in the press. See also, A. N. Winchell, *American Mineralogist*, vol. 14, 1929, p. 125.

TABLE I

DISPERSION LIQUIDS	THERMAL VARIATION LIQUIDS
1. Methylene Iodide	1. Methylene Iodide
2. α -Iodonaphthalene	2. Methylene Iodide+Iodobenzene
3. α -Iodonaphthalene+	3. α -Iodonaphthalene
α -Bromonaphthalene	4. α -Iodonaphthalene+
3a. o-Bromiodobenzene	α -Bromonaphthalene
4. Phenylisothiocyanate	5. o-Bromiodobenzene
5. Iodobenzene	6. Phenylisothiocyanate
6. Bromoform	7. s-Tetrabromoethane
7. o-Toluidine	8. Iodobenzene
8. o-Nitrotoluene	9. Bromoform
9. Propylene Bromide	10. Anilin
10. Methyl Furoate	11. o-Toluidine
11. Methyl Thiocyanate	12. Nitrobenzene
12. Trimethylene Chloride	13. Ethylene Bromide
13. Ethyl Monochloroacetate	14. Propylene Bromide
	15. Pentachloroethane
	16. Methyl Furoate
	17. Methyl Thiocyanate
	18. Isoamylsulphide
	19. Ethyl Dichloroacetate
	20. Ethyl Monochloroacetate

advantage accrued, namely, that it is not often necessary to use more than one liquid to obtain a suitable dispersion curve, that is several readings at different wavelengths. If dispersion is used alone without temperature control then the reading obtained almost never falls on the soda line. Two or more readings in as many liquids are necessary in order to construct the dispersion curve from which the value for the soda line may be read. Such a procedure is laborious. Now if by varying the wavelength the index of liquid and crystal may be made to agree, then by changing the temperature five or ten degrees and thereby changing the index of the liquid, an essentially new medium is obtained. A further change in wavelength once more makes the indices coincide. In this way any number of points on the dispersion curve of the crystal may be obtained all from the same mount. If the crystal itself has high dispersion then the temperature variation alone is useful. Only a few minerals have as high a dispersion as do liquids of the same general index. The entire procedure is based on the fact that almost all liquids have a higher dispersion than crys-

tals and that the temperature coefficient of refraction of liquids is considerable whereas that of most solids is negligible. In order to enhance the value of the method an elaborate search was conducted for suitable liquids—that is liquids having both high dispersion and high temperature coefficients of refraction. These are listed in Table I which includes also a list for use of temperature variation only.

The work is best done in a dark room where there is no light from extraneous sources; also the sensitivity of the eye is slightly greater in a dark room. The apparatus needed in addition to the microscope includes a monochromator, a refractometer, an arc light, a water circulation system with thermometers and a source supply of hot and cold water. The mount is made on a cell through which the water flows. The same water passes through the refractometer on which the same immersion liquid is placed. Light from the monochromator is used to illuminate both the microscope and the refractometer. The temperature is controlled by varying the mixture of hot and cold water—both the microscope cell and the refractometer change temperature together. The wavelength of light is changed by adjusting a simple screw on the monochromator. When the proper conditions are obtained on the microscope then the light is quickly transferred to the refractometer by a mirror rotation and the index is read and corrected by the correction curve supplied with the instrument.

An essential feature of the preliminary paper is the set of liquids described in it. At that time the exact critical data of all the liquids were not known, but measurements were made on the purest liquids on hand. Further work has included careful distillation of fresh supplies and optical data are given for these definite fractions. Most of the liquids were obtained from the Eastman Kodak Co. and these were redistilled. No difficulty has been encountered thus far in duplicating with quite satisfactory accuracy any of the liquids listed. A difference of as much even as one or two in the third place of decimals in the refractive index is not serious provided the liquid used is a real narrow fraction. But closer limits than this are not difficult to obtain. If the dispersion and thermal coefficient of refraction of two supplies of liquid are not the same it does not prejudice the results provided a refractometer is used

TABLE II.

No.	Liquid	Boiling Point °C	mm.	Tempera- ture	$N_F - N_G$	N_D 10°C 25°C 50°C	N_G 10°C 25°C 50°C	N_F 10°C 25°C 50°C
1.	Methylene Iodide	80	15	Coef. .00068	at 25°C .0369	(1) 1.7474	(2) 1.7375	(3) 1.7744
2.	α -Iodonaphthalene	160	14	.000470	.0373	(4) 1.7060	(6) 1.6962	(6) 1.7340
3.	α -Iodonaphthalene + α -Bromonaphthalene	—	—	.000468	.0338	(7) 1.6815	(7) 1.6720	(7) 1.7056
4.	Phenylisothiocyanate	76	4.2	.000562	.0353	1.6555	1.6458	1.6817
5.	Iodobenzene	69-71	16.5	.000570	.0247	1.6252	1.6179	1.6436
6.	Bromobenzene	147.5	73.6	.000507	.0181	1.6029	1.5970	1.6159
7.	α -Toluidine	87-88.5	17	.000507	.0225	1.5768	1.5700	1.5934
8.	α -Nitrotoluene	220	74.0	.000492	.0228	1.5510	1.5435	1.5682
9.	Propylene Bromide	31.5-32	11.5	.000540	.0126	1.5240	1.5198	1.5440
10.	Methyl Furoate	66-66.5	9	.000451	.0166	1.4914	1.4850	1.5035
11.	Methyl Thiocyanate	33.5-34	17	.000538	.0111	1.4735	1.4659	1.4814
12.	Trimethylene Chloride	119.5	73.5	.000490	.0082	1.4534	1.4459	1.4586
13.	Ethyl Monochloroacetate	141.5	74.4	.000474	.0078	1.4265	1.4190	1.4309
3a.	α -Bromotolobenzene	305	74.0	.000517	.0261	1.6677	1.6600	1.6867
7.	<i>s</i> -Tetrabromoethane	139-42	32	.000528	.0177	1.6421	1.6340	1.6785
10.	Anilin	67	4.5	.000523	.0245	1.5911	1.5830	1.6098
12.	Nitrobenzene	66, 68-68.5	5	.000480	.0245	1.5567	1.5490	1.5698
13.	Ethylene Bromide	130	73.3	.000556	.0188	1.5435	1.5350	1.5375
15.	Pentachloroethane	158-60	74.0	.000486	.0098	1.5081	1.5009	1.5213
18.	Isoamylsulphide	80-83	10	.000449	.0091	1.4576	1.4510	1.4886
19.	Ethyl dichloroacetate	84.5	59	.000471	.0084	1.4409	1.4339	1.4396

(1) Calculated from Pulfrich reading at 17°C and the helium wavelength.

(2) Calculated from Pulfrich reading at 17°C and the red hydrogen line.

(3) Calculated from Pulfrich reading at 25°C and the blue hydrogen line.

(4) Calculated from Pulfrich reading at 9°C and the yellow helium wavelength.

(5) Calculated from Pulfrich reading for the yellow helium wavelength.

(6) Calculated from Pulfrich reading for 9°C.

(7) Calculated from Abbé reading for 15°C.

All other readings than those for liquids one and two were made on a Bausch & Lomb direct reading monochromator. The maximum error of these index figures is believed to be $\pm .0002$.
 light source was a Bausch & Lomb Abbé refractometer at controlled temperature and wavelength of light. The

in the system and the same liquid is mounted on the refractometer and on the microscope cell. For the set of liquids using temperature change alone without a refractometer the thermal coefficient of the liquids must be known. The data offered in Table I are those of entirely new distillates from those published in the preliminary paper. One change has been made—liquid no. 3 has been found objectionable in that crystals of dibromonaphthalene separate out in time. A mixture of α -Bromonaphthalene + α -Iodonaphthalene is now used. No suitable substitute has yet been found for bromoform; it therefore is still included.

Critical data for the liquids used are given in Table II. The curves for these data are shown in Figures 1 and 2.

The value of accurate temperature control has been stressed especially. Under ideal conditions the temperature of the mount and of the liquid on the refractometer will be exactly the same. Although this is impossible yet it has been found possible to make them quite reasonably close. The greatest difference between the thermometers which precede and follow the instruments is found at the highest temperatures. For the ordinary run of work one degree difference in temperature is usual, but by increasing the flow slightly this can be cut down to one-half a degree difference. For the liquids used the difference in index for each degree Centigrade ranges from .00045 to .00070. The error due to this cause is practically negligible for the ordinary run of determinative work and for careful work can be cut down to a maximum of .0003 under the most unfavorable circumstances. At lower temperatures at which the operator usually prefers to work this error is less. I would caution the operator to check the thermometers against each other for unless the two have been carefully selected they will not agree. Usually when it is necessary to resort to high temperatures (50°–60°C) it is also possible to check the result on the next liquid at a low temperature. The two results should be close and a very close figure can be estimated between them.

Dr. H. E. Merwin raised the very pertinent question⁴—“How closely does the temperature of the liquid on the microscope cell agree with the indicated temperature of the thermometers?” At his suggestion I employed the method of Ashton and Taylor⁵ to check the temperatures. The “cold junction” of a thermocouple was placed on the refractometer and the “hot” junction was placed

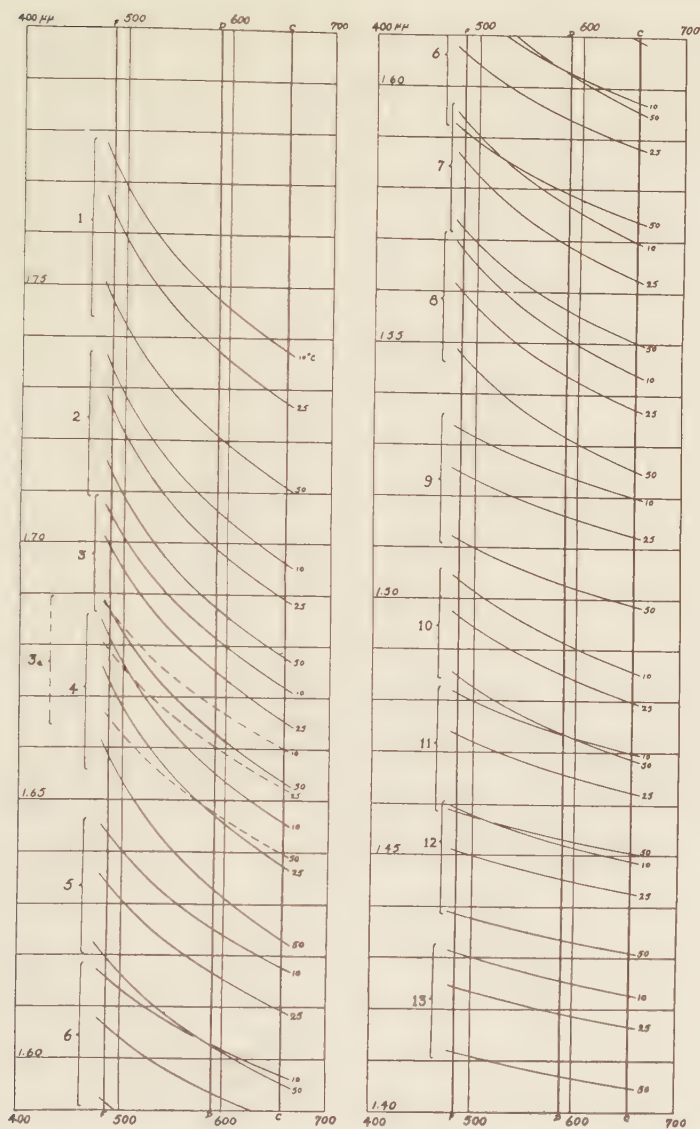


FIG. 1. Graphical representation of the dispersion and temperature coefficients of refraction of the set of liquids in which both these properties are used. This graph is intended to be used as a guide in selecting the proper liquid to be used in a determinative procedure.

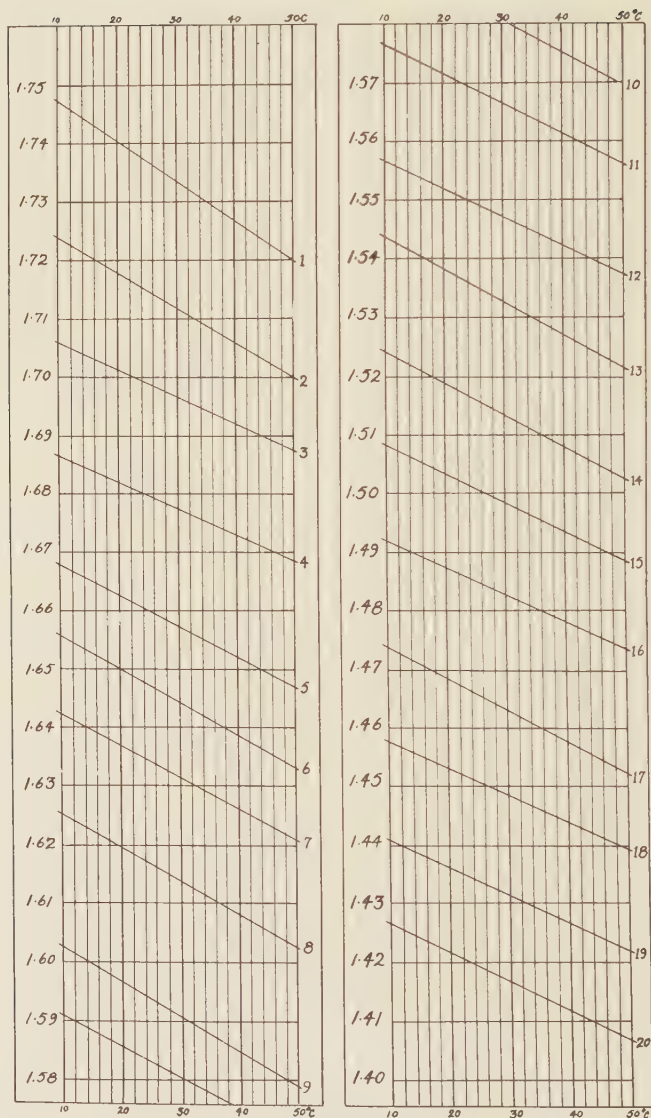


FIG. 2. Graphical representation of the index range of a set of liquids for temperature variation only. This set is intended to obviate the necessity of mixing liquids and to reduce the number of mounts necessary in a procedure, at the same time avoiding the expense of a refractometer and a monochromator.

on the water cell. A Hoskins pyrometer type "H. A." was used in which one half millivolt was subdivided and the instrument calibrated to read accurately to one half degree Centigrade and by interpolation to one quarter degree. In this way the temperature difference between the liquid on the refractometer and that on the water cell was read:

Room		temperature 27°C	
At 28.5°C	after	2 minutes difference =	0°
38.5°C		1	1°
		6	$\frac{1}{4}$ °
49.5°C		2	$1\frac{1}{4}$ °
		4	1°
		6	1°

Empirical tests indicate that these differences are quite general, and allowance can be made for them for accurate work.

A change in temperature of fifty degrees will affect the index of the glass in the refractometer. The relative importance of this source of error is indicated in the following extract from a letter from the Bausch & Lomb Optical Co.

"The temperature coefficient of glass of the type used is given as 0.0000066 t. This would mean a total index change between 10° and 50° of 0.00026. Since the scale is computed for N_d of approximately 25° this error would be slightly reduced, in that it would become plus and minus. Such a variation in index of the prism would mean about the same index change in the sample. I should say that it would never be over 0.0002 and seldom as high as that. You will understand that in this statement I am assuming a glass type, rather than the particular glass in the instrument. The temperature constants of the particular glass have never been determined."

The maximum error then on a single reading should be $\pm .0005$. Experience indicates that it is usually less. A desired reading at a particular wavelength, usually near $589\mu\mu$, is best made by taking an average.

I recently received for examination from Bausch & Lomb Optical Co. a first trial model of the set-up as they propose to make it. The accompanying photographs (Figs. 3a and 3b) show

⁴ Personal communication.

⁵ F. W. Ashton and W. C. Taylor, A Precision Method for Measuring Temperature of Refractive Index Liquids. *Am. Mineralogist*, vol. p.13, 44.

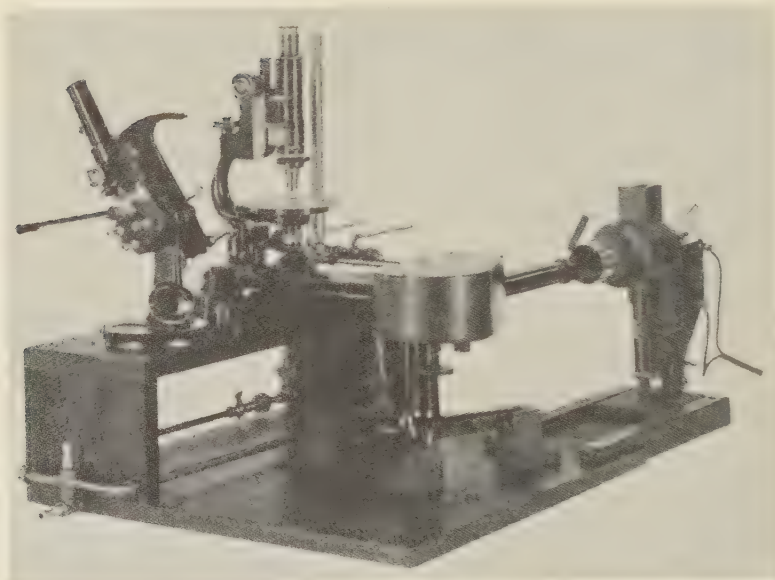


FIG. 3a. Bausch and Lomb apparatus complete but without the most recent changes.

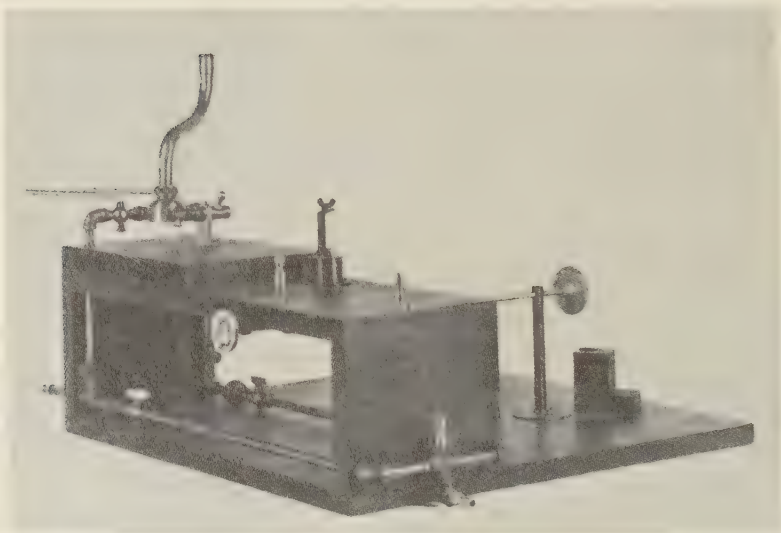


FIG. 3b. Base of Bausch and Lomb apparatus with the final improvements added.



FIG. 4. Double variation apparatus being used in the Geological Laboratories at Madison.



FIG. 5. Double variation apparatus being used in the Geological Laboratories at Madison.

the set-up including a few simple suggested changes recommended after the apparatus was carefully tried out in our laboratory. Bausch & Lomb instruments are used throughout.

Our own equipment was remodeled to include all modifications deemed advisable on the basis of our own experience and our observations of the Bausch & Lomb arrangement. Bausch & Lomb made some very real improvements over our original model after which theirs was designed. Two complete units are shown in Figures 4 and 5. The apparatus of Figure 4 is Bausch and Lomb equipment. For this purpose it is well to use an enclosed arc made by both companies, in which the air circulation is staggered to prevent the diffusion of light in the room. The apparatus of Figure 5 is Leitz except for the refractometer which is a Spencer model. Both Bausch & Lomb and Spencer refractometers are found quite satisfactory. The difference in mechanical manipulation is great enough to justify individual personal preference on the basis of these features. Correction curves for use with monochromatic light are supplied by both companies.⁶

The Leitz monochromator has been used with satisfaction, but I feel that the following features of the Bausch & Lomb monochromator make it more desirable for this purpose. The Bausch & Lomb instrument is direct reading in $\mu\mu$ on a four inch drum. The Leitz instrument is much more compact but is not direct reading. The scale on it is divided into one hundred units—(the F line on the instrument in Figure 5 reads 51.5 and the C line 24.2) on a 1 1/4 inch drum; readings are therefore much more accurate on the Bausch & Lomb instrument. The Bausch & Lomb instrument also has a small prism in the collimator tube for securing a comparative spectrum. The outstanding disadvantage of the Bausch & Lomb monochromator, as I see it, is its lack of dirigibility. The Leitz instrument is mounted on a very conveniently adjustable hinge which gives it a general utility not possessed by the other. The Leitz monochromator also has a neatly adjustable collimator prism which greatly facilitates illumination of the instrument. These features are pointed out in the hope of assisting those interested who are not familiar with

⁶ There are other makes of Abbé refractometer which doubtless will serve quite well; a Pulfrich is not desirable for this type of work.

both instruments, to make a selection. The importance of the monochromator for the purpose of this paper cannot be over emphasized. The monochromator should be carefully checked before being used, either with spectrum tubes or standard flames. If the Bausch & Lomb base is used then only Bausch & Lomb instruments can be made to fit without modifying the base. I feel, however, that the complete Bausch & Lomb unit is very satisfactory.

I have been asked if interference figures can be obtained with the cell in place. They cannot by any method known to me. Optic axis grains are not difficult to recognize and optic normal grains though not so easy to recognize are less often needed. To measure n_p and n_o I believe the standard method is best—namely to choose the grains of highest interference colors in the 45° position and measure a few of them, selecting extreme values. It is to overcome this recognized limitation that I have spent considerable time attempting to adapt the universal stage to this purpose. If successful it should offer an extremely rapid, accurate and generally satisfactory procedure.

In our original apparatus tap water was used directly from the mains. This has been abandoned entirely because of the dissolved air that is released from the hot water immediately on its passing the valve. Water from the mains is now run into two tanks—one hot, one cold—and from these the water for the apparatus is drawn. An overflow from the tanks keeps them at a temperature equivalent to the source temperature. The dissolved air is mostly removed while in the tanks. A bubble trap in the system removes almost all the remaining bubbles. The cell developed at considerable pains by Mr. J. S. Hipple serves very well indeed to allow the remaining bubbles to pass directly through.

On first turning on the water the flow is usually hampered by bubbles trapped in the refractometer and probably elsewhere. A few sharp taps on the tubing serve to release these and the flow increases greatly. A good flow of water is absolutely essential to good work.

Experience indicates that one of the most common sources of error on the part of students lies in failure to give the refractometer sufficient time to come to temperature. It is best to make a series of readings until two agree. If the mineral substance is

clear and gives a sharp Becke line then the reading on the microscope, after changing the temperature, can usually be made more rapidly than the refractometer can come to temperature. Students who spend more than two or three hours at a time making readings seldom secure reliable results near the end of the period. Their readings sometimes are more than .002 off, due apparently to eye fatigue.

PROBERTITE, A NEW BORATE

ARTHUR S. EAKLE, *University of California.*

The new borate described in this paper occurs as one of the minerals of the kernite deposit¹ in the Kramer District, Kern County, California, and the name "probertite" is proposed for the mineral, in honor of Frank H. Probert, Dean of the Mining College, University of California, to whom the writer is indebted for specimens, photos and notes of its occurrence.

The mineral is prismatic columnar, radiating from sharp centers and appearing as rosettes up to three centimeters in diameter. It occurs as inclusions in the kernite (Fig. 1), in secondary borax (Fig. 2), and in the greenish gray shales which the kernite deposit has apparently replaced, (Figs. 3 and 4). It is of fairly common occurrence in the deposit.

Probertite is colorless, glassy and brittle, crushing into long splinters some of which show prismatic cleavage. No crystal faces are present. $H=3-4$. $Sp. G.=1.91$. It is a double borate of sodium and calcium but is distinctively different from ulexite in having less water and more sodium.

ANALYSIS		RATIOS
Na ₂ O	14.03%	.226
CaO	12.76	.228
B ₂ O ₃	47.80	.675
H ₂ O	24.83	1.378

From the ratios it is evident that the formula for probertite is $Na_2CaB_6O_{11} \cdot 6H_2O$. This is analogous to the formula for hydroboracite $CaMgB_6O_{11} \cdot 6H_2O$. No magnesium minerals have been so far observed in the kernite deposit. In a low Bunsen flame probertite first whitens and then fuses easily and quietly to a clear glassy bead with no intumescence or decrepitation. It is little affected by cold water and very difficultly soluble in hot water. Its solubility is less than for ulexite. Easily soluble in acid solutions.

It crystallizes in the monoclinic system. Biaxial, positive with large 2V angle.

¹ Schaller, W. T., Kernite, a new sodium borate. *Amer. Mineral.*, 1927, vol. 12, p. 24.

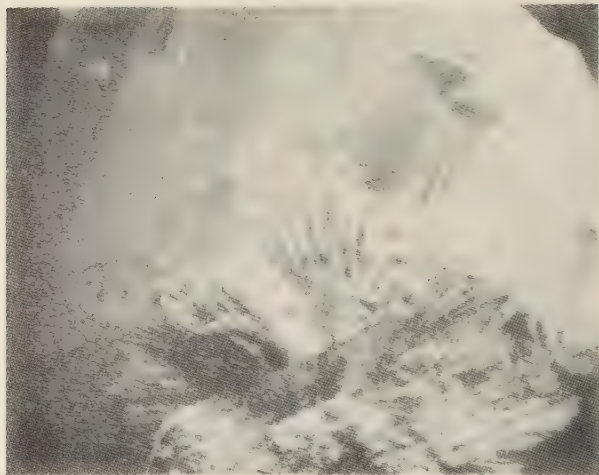


FIG. 1. Probertite in kernite. Magnified 2 diameters.

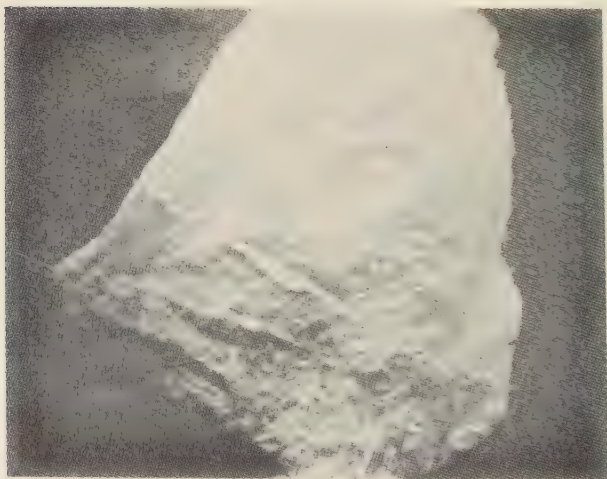


FIG. 2. Probertite in massive borax. Magnified $1\frac{1}{2}$ diameters.



FIG. 3. Probertite hemisphere enclosed in shale. Magnified $1\frac{1}{2}$ diameters.

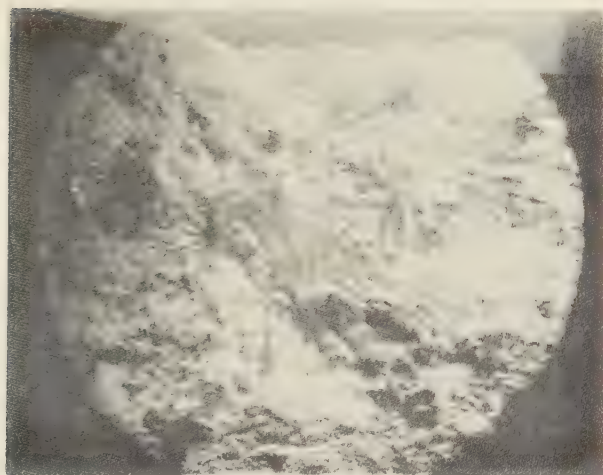


FIG. 4. Probertite rosettes in shale. $\frac{2}{3}$ Natural size.

Indices— $\alpha=1.515$; $\beta=1.520$; $\gamma=1.538$. Z to $c=12^\circ$.

These indices lie between those of ulexite and hydroboracite:

Ulexite $\alpha=1.491$; $\beta=1.504$; $\gamma=1.520$.

Hydroboracite $\alpha=1.517$; $\beta=1.534$; $\gamma=1.565$.

Kernite and probertite are the primary borates of the deposit and the crystallization of the probertite has preceded or has been simultaneous with the crystallization of the kernite. The deposit is a lode-like replacement deposit in Upper or Middle Miocene-Tertiary beds. These two exceptional borates are evidently products of fumarolic or hot spring conditions in which the temperature was the important factor influencing the crystallization of these two lower hydrates, as the primary minerals of the deposit, instead of the usual and common borax and ulexite. The presence of realgar, orpiment and stibnite as associates of the kernite confirm this view of fumarolic action.

Borax is very common in fresh glassy masses and in white altered veins, but is wholly secondary and a result of the alteration of the kernite. Ulexite is also common in divergent masses, and in veins like satin-spar, but its origin represents a later crystallization from borate solutions at ordinary temperatures.

INTERPRETATION OF CONCENTRIC TEXTURES AT COLQUIJRCA, PERU

H. E. MCKINSTRY, *Cambridge, Mass.*

Apropos of recent publications on the interpretation of micro-textures in terms of the sequence of minerals¹ it may be of interest to describe an unusual case of concentric arrangement of sulphides. The illustrations show stromeyerite, galena, sphalerite and chalcopyrite in textures which might be (and by a very able investigator were) interpreted as meaning that stromeyerite was deposited both before and after galena and the other minerals, hence was of the same general age as the galena and consequently hypogene. The writer, who collected the specimens at Colquijirca, Peru, suspected from field evidence that the stromeyerite was supergene and later had the privilege of studying the material in detail in the Harvard Laboratory. In polished sections the texture was very perplexing and it was not until hand specimens were studied under the binocular microscope that the proper interpretation suggested itself.

In a number of specimens, galena was found to be deposited on tennantite crystals, partly replacing the tennantite and partly growing upon it. Sphalerite forms botryoidal crusts upon the galena and at times there is still another crust of galena upon the sphalerite (Fig. 1 (1)). Chalcopyrite is later than all of these minerals, replacing them and forming crusts upon them but is particularly active in its replacement of galena. Not infrequently chalcopyrite is seen to have broken through the band of sphalerite and attacked the underlying galena as shown graphically in Fig. 1, (2).

The latest sulphide is stromeyerite. It fills the vugs which were lined by chalcopyrite but in replacing shows a strong preference for tennantite. Some specimens show tennantite cut by veinlets of stromeyerite and others contain remnants in all stages of replacement. Frequently stromeyerite leaves the outer crust of chalcopyrite and the other minerals almost untouched and passes inward to replace the core of tennantite (see Fig. 1, (3)).

Wherever stromeyerite is abundant it is accompanied by evidence of etching and corrosion of the other sulphides as if by acid.

¹ Fairbanks E. E., *The Laboratory Investigation of Ores*; McGraw-Hill Book Co., *New York* 1928, p. 132 (Colony); p. 147 (Newhouse).

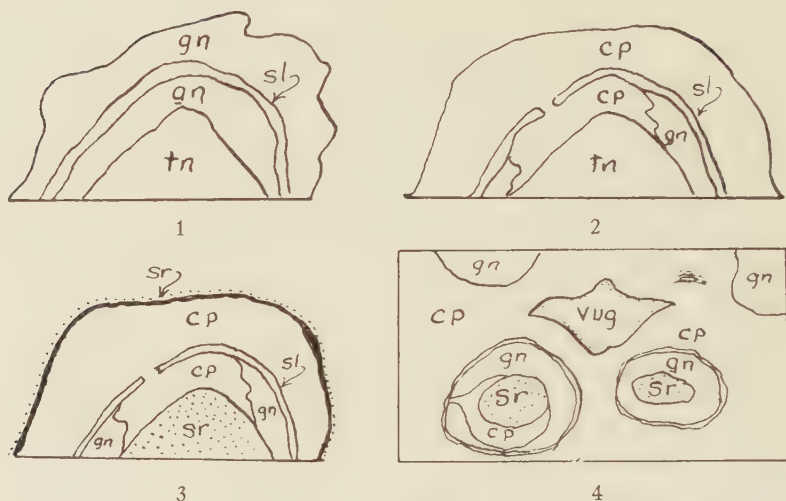


FIG. 1. Successive stages in the development of inherited concentric textures.

(1) Crystal of tennantite (tn) protruding into vug coated by alternate depositions of galena (gn) and sphalerite (sl).

(2) Selective replacement of galena by chalcopyrite (cp).

(3) Deposition of stromeyerite (sr) upon chalcopyrite and also replacing cores of tennantite.

(4) A group of such coated and replaced crystals as they would appear in polished section.

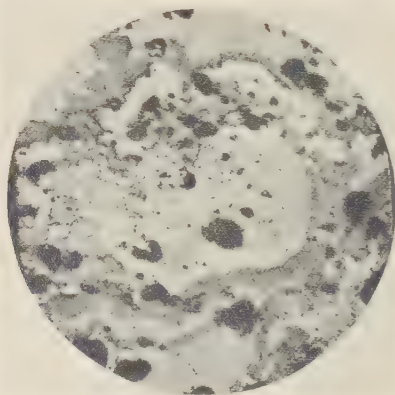


FIG. 2. In the center are tennantite and galena (in part intergrown with pierceite?) surrounded by a white rim of galena. Outside this are two somewhat discontinuous rims of sphalerite (dark gray). The space between the two sphalerite rims is occupied in part by galena and in part by chalcopyrite, the space outside the outer rim being occupied by chalcopyrite.

x60 (Photo by M. N. Short and R. W. Goranson).

With the stromeyerite, native silver is almost universally associated, cutting it in veinlets and growing from it in wires within the vugs. The attack by the corrosion which accompanies the development of stromeyerite may be likened, to use an unpleasant analogy, to the decay of a tooth where the enamel is left mainly intact but the core of less resistant material succumbs.

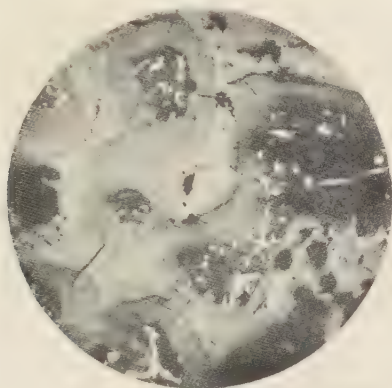


FIG. 3. Cores of stromeyerite (which has totally replaced tennantite) and native silver, surrounded first by galena and then by chalcopyrite. Vugs like the one represented by a black spot on the right ordinarily contain stromeyerite and silver. x60 (Photo by M. N. Short and R. W. Goranson).

While all of the stages described are not necessarily found in any one specimen and hence cannot well be shown in any one illustration, examples of all stages of the process have been observed. The facts that stromeyerite is the latest of the sulphides, is accompanied by etching suggesting acid attack and is always associated with native silver, are perfectly in accord with the true field observation that the stromeyerite stage of deposition is absent in those parts of the mine which are least favorably located with respect to supergene enrichment and which show correspondingly lower content in silver.

SUMMARY

In this particular case, an example of concentric texture was shown to have originated not from simple rhythmic precipitation in a gel nor by simple successive stages of deposition but involved highly selective replacement at two stages, the last of which, at least, was supergene.

NOTES AND NEWS

VASHEGYITE AND BARRANDITE IN NEVADA

H. G. CLINTON, *Manhattan, Nevada.*

The aluminum phosphates vashegyite ($4 \text{ Al}_2\text{O}_3 \cdot 3 \text{ P}_2\text{O}_5 \cdot 30 \text{ H}_2\text{O}$), barrandite ($(\text{Fe}, \text{Al})_2\text{O}_3 \cdot \text{P}_2\text{O}_5 \cdot 4 \text{ H}_2\text{O}$), and utahlite ($\text{Al}_2\text{O}_3 \cdot \text{P}_2\text{O}_5 \cdot 4 \text{ H}_2\text{O}$), are all found in the Vashegyite Gem Mine at Manhattan, Nevada. These phosphates are of rather rare occurrence. Vashegyite has been reported from only one other locality, Vashegy, Hungary; barrandite is found only at Příbram, Bohemia; while utahlite is found elsewhere only in Tooele County, Utah.

The Vashegyite Gem Mine is an open cut in altered slate near a porphyry contact. The vein is two to three inches wide and occupies a fracture that strikes in a northeast-southwest direction across the bedded formation and dips to the north. The main mineral in the vein is variscite, variety utahlite, which occurs in lenticular masses. The utahlite is often "frozen" to one or both walls, but is divided in the vein into various shapes by fibrous crusts and loose aggregates of aragonite. In color the utahlite varies from dark emerald green to light pea green. It may be also yellow, purple, blue, brown or pink. Much of this is gem material and is cut by lapidaries at Idar, Germany.

The vashegyite, which is white or yellowish white in color, occurs as a filling in the vein at the east end of the fracture where a quartz ledge forms the foot wall and extends downward for a distance of about three feet where it passes over into utahlite. The utahlite in turn grades into turquoise by the addition of fourteen per cent aluminum oxide and the loss of about twelve per cent of phosphorus pentoxide.

At one point the green opal distributed between the layers of vashegyite gave place to blue opal, which in turn became brown as it approached a seam of barrandite. It was not of gem quality, however, and disappeared with the vashegyite.

Lying in a horizontal position in the north wall is a fissure, three eighths of an inch thick filled with barrandite, while a massive vertical ledge of white quartz, twelve inches in thickness, is cut by the seam of barrandite. On the south side of this quartz ledge the barrandite is green in color, while that in the seam proper as well as the material to the north is yellow, the outer edges grading over to red.¹ Most of this massive quartz ledge above the barrandite is replaced by yellow barrandite and associated with this mineral are brown crystals of jarosite² ($\text{K}_2\text{O} \cdot 3 \text{ Fe}_2\text{O}_3 \cdot 4 \text{ SO}_3 \cdot 6 \text{ H}_2\text{O}$). Probably turgite will also be found with the jarosite but it has not yet been reported. The mineral dahllite is found in lustrous silver white crusts in the cavities and cracks in the quartz of the altered slate, also in the barrandite.

The open cut is now twenty feet long by eight feet wide, with a depth of, twelve feet at the deepest place. Near at hand is a large intrusion of rhyolite. The metamorphism due to heat, pressure, and the chemical action of solutions have brought about the following changes:

Mudstones have been altered to pink and cream colored, silky schists.

Slate replaced by barrandite and by blue opal.

White quartz replaced by barrandite. Slate altered to black jasper.

Quartz and schists altered to limonite and sericite.

¹ Described by Earl V. Shannon, *Am. Mineral.*, vol. 8, p. 182, 1923.

² Found by W. F. Foshag.

The minerals thus far found in this open cut are: utahlite in five colors; barrandite, green, red and yellow; turquoise; opal, blue, green, brown; vashegyite; dahllite; jarosite; quartz; francolite; limonite; manganese oxide; chrysocolla; jasper; chalcedony; metacalcio wardite; chlorophanite; sericite; calcite; and aragonite.

PERCENTAGES FOR COMPARISON

	Al ₂ O ₃	P ₂ O ₅	CO ₂	Fe ₂ O ₃	CaO	Cl	H ₂ O
Barrandite	12.5	40.7	..	26.2	20.6
Utahlite	32.3	44.9	22.8
Turquoise	46.8	32.6	20.6
Vashegyite	29.7	30.9	39.2
Dahllite	..	39.0	6.0	..	53.7	..	1.3
Francolite	..	41.0	53.8	6.8	0.0

PARAGENESIS OF THE DEPOSIT

The fracture resulted from a displacement in the Black Mommoth Hill. The bedded formations in the north wall were contorted and the layers separated in places by an intrusion of rhyolite. The quartz ledge was formed prior to the tilting and fracture. The hot aqueous solutions entering with the rhyolite dissolved the aluminum phosphates in their passage through the surrounding limestones, slates and schists. After entering this fracture the solutions probably remained practically stationary except for small amounts that slowly seeped into the intersecting fissures. Elevated temperatures were no doubt maintained for a very long period by the massive intrusive which cooled very slowly.

The solutions in contact with the quartz ledge of the foot wall reacted and dissolved some of the silica which later separated as opal after the aluminum and phosphorus had deposited as vashegyite. Neither of these two minerals was deposited in a continuous mass over the allotted space, but first one, then the other would be formed covering small areas from two to four square inches each. The thickness of the layers varies from that of a sheet of paper to a quarter of an inch, with opal usually, but not always, constituting from four to ten times the volume of the vashegyite. It is also to be noted that the same solutions that deposited white vashegyite gave rise to green, blue and brown opal. The coloring matter in the opal came in part from the copper mineral chrysocolla, and in part from the barrandite. As the fissure containing these minerals stands nearly vertical ample proof is furnished that the solutions at this point were of a jelly-like consistency.

None of the solutions in the fissure removed from the influence of colloidal silica formed vashegyite. Instead, in the absence of silica, the aluminum phosphates formed utahlite and turquoise (see table showing percentages).

In places the walls of the fissure are altered to black jasper. Solutions absorbing a portion of this black pigment and combining with the green of the chrysocolla gave a very beautiful emerald green color to the utahlite near the walls. Intermingled with the green are spots of velvety brown with streaks of yellow, pink and purple.

Near the western end of the open cut is a zone of sheeting consisting of several fractures now filled with quartz which crosses the fissure at right angles. The

circulating solutions entering these fractures brought in some calcium and chlorine. These newly added elements did not combine with the alumina, but after all the alumina and part of the phosphorus pentoxide had been thrown out of solutions as utahlite, the calcium and chlorine united with the remaining phosphorus pentoxide to form francolite.

The francolite is often in the center of a lens of utahlite, but it also occurs nearly pure and at times contains isolated spots of the new mineral metacalciovardite which has not yet been described.

A fissure at the east end of the deposit permitted a portion of the solutions to pass through some limonite from which they extracted enough iron to form barrandite. The mineral dahllite was not formed from solutions passing through the main fissure but from solutions originating near the surface.

A study of this deposit indicates how minute changes may produce a number of different minerals from the same solution. If this main fissure had been two inches farther north, so as to have missed the quartz ledge of the foot-wall, neither opal nor vashegyite would have been formed. Transposing the body of limonite in the north wall to the position occupied by the quartz ledge would have filled the main fissure with barrandite. Had the zone of sheeting been two feet farther west the chlorine-calcium solutions could not have entered the fracture and there would have been no francolite.

Following the period of formation this deposit was overlain by not less than five hundred feet of limestones, slates and schists, all of which have been eroded. It is expected that other rare or new minerals will be found with greater depth.

Mr. A. W. Quinn has been appointed instructor of mineralogy and petrography in the department of geology of Brown University. Mr. Quinn takes the place of Dr. M. E. Hurst who has joined the staff of the Ontario Bureau of Mines.

Mr. James H. C. Martens has accepted the position of assistant professor of mineralogy and petrography in the department of geology at West Virginia University.

Two offices have recently been opened by E. Leitz, Inc., where a complete stock of Leitz products will be on display, one in the Peoples Gas Building, 122 South Michigan Ave., Chicago, Illinois, and the other in the Investment Building, Washington, D. C.

Dr. George Perkins Merrill, head curator of geology in the U. S. National Museum, died suddenly of a heart attack in Auburn, Maine, on August 15, 1929. He was an indefatigable worker and during his long and active career contributed more than 150 papers on mineralogical and geological subjects, about 60 of these pertain to meteorites. His larger contributions include such well known works as: *Stones for Building and Decoration*; *Rocks, Rockweathering and Soils*; *The Non-metallic Minerals—their Occurrence and Uses*; *Hand-book and Descriptive Catalogue of the Collections of Gems and Precious Stones in the U. S. National Museum*; *Contributions to the History of American Geology*; *The First One Hundred Years of American Geology*; *History of American State Geological and Natural History Surveys*; and *Minerals from Earth and Sky*.

Attention is again called to the necessity of sending at once to the Secretary, Prof. Frank R. Van Horn, Case School of Applied Science, Cleveland, Ohio, titles of papers to be presented before the annual meeting. A preliminary list of titles will appear in the December issue of the Journal.

BOOK REVIEWS

THE MINERAL WEALTH OF THE BLACK HILLS. JOSEPH P. CONNOLLY AND CLEOPHAS C. O'HARRA. Bulletin No. 16, SOUTH DAKOTA SCHOOL OF MINES. 418 pages+64 plates. Published May 1929.

Many papers have been published from time to time on the economic geology and mineralogy of the Black Hills region but bulletin No. 16 is the only recent publication which covers all phases of the subject.

When the mineral wealth of this region is mentioned the casual reader, no doubt, has visions of rich gold deposits and also possibly of important lithium minerals. These impressions are but natural as the Black Hills produce annually more than one eighth of the gold mined in the United States and since 1875 the value of this metal alone has exceeded \$274,000,000. Also the largest lithium producing mine in the World is located here and the size of the spodumene crystals has not been equalled elsewhere. (One crystal had a length of 42 ft. and a cross section of 3×6 ft. Theoretically such a crystal should yield about 90 tons of spodumene but it was so badly weathered that only 37 tons of commercial spodumene were obtained. Dr. Schaller has called attention to a still longer crystal, one measuring 47 feet in length).

This, however, is only part of the story. Over one hundred different species including both primary and secondary minerals have been listed as occurring in the pegmatites of the Black Hills, while the value of the mineral production since mining began fifty-four years ago has totaled approximately \$335,000,000. Aside from gold, in the period from 1875-1928, the value of the production of silver, tungsten, mica, cement, gypsum, building stone, sand and gravel, clays, petroleum and natural gas, and coal has been, in each case, in excess of \$1,000,000. Many other minerals of economic importance have been recovered but the values have been less than those mentioned above.

The bulletin is written in a semi-popular style and covers not only the geology and mineral production of the region but also touches upon such phases of petrology as the origin of the pegmatites. The magmatic crystallization theory and the hydrothermal replacement theory are discussed with special reference to their application to the dikes of the Black Hills.

The bulletin contains 64 plates as glazed inserts and these contribute much to the interest and attractiveness of the volume. A copy of bulletin No. 16 can be purchased for the nominal sum of eighty cents, plus postage, by addressing the South Dakota School of Mines at Rapid City, South Dakota.

W. F. H.

PROCEEDINGS OF SOCIETIES

PHILADELPHIA MINERALOGICAL SOCIETY

A stated meeting of the Philadelphia Mineralogical Society was held September 5, 1929, with the President, Mr. Trudell, in the chair. There were 36 persons present including 6 visitors. In the absence of Mr. Gordon, Mr. Strock acted as secretary.

Upon favorable recommendation of the Council Messrs. Allen Bernheimer, Morris Nasselau and Robert Williams were elected Junior Members. Nominations for officers were deferred until the October meeting.

The program of the evening, accounts of summer trips, indicated some very active mineral collecting by the various members during the past summer. Mr. Vanartsdalen reported on a trip which included Lee, Massachusetts, Bedford, Herkimer and Tilly Foster, New York. Fine Tremolite was obtained at Lee and fresh mineralogical material was obtainable at Tilly Foster from the dumps which are being removed. Mr. Poole described a trip to the Finger Lakes region, N. Y. Mr. Cienkowski reported briefly upon a 7,000 mile automobile trip which included Hot Springs and Magnet Cove, Arkansas; Joplin, Missouri; Keokuk, Iowa; and various localities in North Carolina, Tennessee, New England and Nova Scotia. Messrs. Schwan and Bernheimer accompanied him on the southern portion of the trip while Messrs. Geunst and Reinitz were with him in New England and Nova Scotia.

Mr. Cajori reported on an extensive trip to Europe where he visited museums in London, Paris, Munich and Edinburgh. He also described the present status of several mineral localities in the Austrian Tyrol, Leadhill district of Scotland and a new locality near Dublin, Ireland. He also gave an account of the diamond cutting industry in Amsterdam. Mr. Benjamin Shoemaker, 3rd, told of a hunting trip to Africa, through the Belgian Congo and Rhodesia. Mr. Leavitt visited several localities in Maine. Messrs. Biernbaum and Strock gave an outline of a trip taken by them to Nova Scotia which will be presented more fully at the October meeting. Local trips were reported by Messrs. Poole, Knabe, Loebel, Strock and others.

LESTER W. STROCK, *Sec'y, Pro-tem.*

NEW MINERAL NAMES

Buszite

E. STEINWACHS: Buszit, ein neues Mineral der ditrigonal-bipyramidal Klasse; von Khanin Südwestafrika. (Buszite, a new mineral of the ditrigonal-bipyramidal class; from Khan, Southwest Africa.) *Centr. Mineral., Geol., Abt. A.*, 1929, pp. 202-205.

NAME: In honor of K. Busz, German mineralogist.

CHEMICAL PROPERTIES: A silicate of rare elements, chiefly neodymium, praseodymium, erbium and europium. Slightly soluble in hydrochloric or sulphuric acids but soluble with the evolution of a gas in perchloric acid. Absorption spectra show the presence of Nd, Pr, Er and Eu.

CRYSTALLOGRAPHIC PROPERTIES: Trigonal, ditrigonal-bipyramidal. $a:c = 1:1.1792$. Forms: $c(0001)$, $R(10\bar{1}1)$, $m(1120)$, $p(10\bar{1}0)$, $s(41\bar{5}0)$. Habit prismatic with prominent base, prism zone striated.

PHYSICAL AND OPTICAL PROPERTIES: Color yellowish red-brown (cinnamon), transparent and yellow in thin splinters. Luster resinous. n about 1.72. Birefringence strong. $H=5.5$. Sp. Gr. 4.977 (contains small included beryl crystals).

OCCURRENCE: As a single crystal with attached beryl from Khan, Southwest Africa.

W. F. FOSHAG

Bialite

H. BUTTGENBACH: Note sur la Bialite, nouveau mineral. (Note on bialite, a new mineral.) *Ann. Soc. Geol. Belg. Publications spec. relat. Congo belge. Année 1927-1928*, pp. 3-9, 1929.

NAME: In honor of Lucien Bia, Belgian explorer of the Belgian Congo.

CHEMICAL PROPERTIES: A hydrous phosphate of lime and magnesia. No analysis is given.

CRYSTALLOGRAPHIC PROPERTIES: Small tufted needles, scarcely 3 mm. long. Cleavage parallel to the prominent face.

PHYSICAL AND OPTICAL PROPERTIES: Color white, luster nacreous. Biaxial, positive. Bx_a = length of the crystal. X is across the plates, Y through the plates, Z along the prismatic direction. β (or α) = 1.525, γ greater than 1.541 and less than 1.546. Cleavage face shows birefringence of 0.00205.

OCCURRENCE: Found on a brown, opaque phosphatic rock from Mushishimano, Katanga.

DISCUSSION: Believed to be similar to tavistockite and perhaps a magnesian analogue of that mineral.

W. F. F.

Mangandiaspore

K. CHUDOBA: Über "Mangandiaspor," und Manganophyll von Postmasburg (Griqualand—West, Südafrika). (Mangandiaspor and manganophyllite from Postmasburg, Griqualand—West, South Africa.) *Centr. Mineral., Geol., Abt. A.*, pp. 11-18, 1929.

NAME: A diaspor with appreciable manganese content.

CHEMICAL PROPERTIES: A manganiferous oxide of alumina. Analysis: SiO_2 0.11, Al_2O_3 78.58, Fe_2O_3 1.96, Mn_2O_3 4.32, CaO tr., H_2O 14.65; sum 99.62.

CRYSTALLOGRAPHIC PROPERTIES: Habit prismatic, flattened parallel to (010). Prism zone vertically striated. Forms: (010), (210), (120). Crystals up to 8 cm. in size.

PHYSICAL AND OPTICAL PROPERTIES: Color rose to dark red. Optical properties like those of normal diaspor.

$\alpha=1.7023$, $\beta=1.7219$, $\gamma=1.7502$. Sp. Gr. 3.328.

OCCURRENCE: Found as lenses and layers exceeding several centimeters in thickness in coarse crystalline manganese ore. Reticulated to radial in structure. Resembles rhodonite.

W. F. F.

Takizolite

S. IMORI AND J. YOSHIMURA: A pink Kaolin, and Ruthenium as a minor Constituent of the Tanokami Kaolins. *Bull. Chem. Soc. Japan*, 4, pp. 1-5, 1929.

NAME: From a local Japanese collector, Takizo Ueno.

CHEMICAL PROPERTIES: Hydrous silicate of alumina. $2\text{Al}_2\text{O}_3 \cdot 7\text{SiO}_2 \cdot 7\text{H}_2\text{O}$. Analysis: SiO_2 53.91, Al_2O_3 26.28, Fe_2O_3 1.69, FeO 0.26, MnO 0.39, TiO_2 0.03, CaO 0.17, MgO 0.09, rare earth oxides 0.67, K_2O 0.62, Na_2O 1.03, F 0.03, ign. loss 15.60; sum 100.72.

PHYSICAL AND OPTICAL PROPERTIES: Color pink, light brown or flesh colored. Streak white. Birefringent. $n = 1.515 \pm 0.001$. $H = 2.5$.

OCCURRENCE: From an altered granite at Tanokami Hill, Oomi Province, Japan.

W. F. F.

Rinkolite

E. M. BONSHTEDT: Two new minerals of the mosandrite group from Monts Chibines. *Bull. Acad. Sci., U.S.S.R.* [6] 20, p. 1181, 1926; A. E. FERSMAN: *Neues Jahrb. Min., Abt. A.*, 55, 44, 1926. (Cf. *Am. Mineral.*, 11, 295, 1926).

NAME: Because of its similarity to rinkite.

CHEMICAL PROPERTIES: Analysis gave: SiO_2 27.58, TiO_2 11.15, ZrO_2 0.35, Ce_2O_3 18.02, Al_2O_3 1.47, Fe_2O_3 0.99, CaO 24.70, SrO 3.30, Na_2O 6.73, K_2O 0.16, F 5.99, H_2O 1.75. Sum. 102.19. Said to belong to the mosandrite-rinkite group.

PHYSICAL PROPERTIES: Yellow-green bladed monoclinic crystals.

$G = 3.40$. $H = 5$. $\beta = b$. $2V = 45^\circ - 88^\circ$,

$\alpha = 1.643$ (1.662), $\beta = 1.645$ (1.667), $\gamma = 1.651$ (1.681).

Cleavage (100) perfect, (010) good. Feebly pleochroic.

OCCURRENCE: Found at many places in the nephelite-syenites of the Kola Peninsula, north Russia.

J. F. SCHAIRER

Barium-Phlogopite

A variety name for a phlogopite from Mansjo Mt., Sweden, containing 1.28% BaO. [H. VON ECKERMANN, *Tsch. Min. Petr. Mitt.*, 38, 282 and 286, 1925.]

Drewite

A variety of calcium carbonate precipitated from sea-water by bacterial action. Named for G. H. Drew (1881-1913) who described it. [R. M. FIELD, *Carnegie Inst. Washington*, Year-Book No. 18 (for 1919) p. 197; E. M. KINDLE, *Pan.-Amer. Geol.*, 39, 368-9, 1923.]

Ferri-Muscovite

Name given to the ferric molecule, $\text{H}_2\text{KFe}_3^{\text{III}}(\text{SiO}_4)_3$, corresponding to muscovite. [W. WAHL, *Fennia*, 45, No. 20, p. 85, 1925.]

Ferri-Orthoclase

Name given to the ferric molecule, $\text{KFe}_3^{\text{III}}\text{Si}_3\text{O}_8$, corresponding to orthoclase. [W. WAHL, *Fennia*, 45, No. 20, p. 48, 1925.]